

# Structural Phase Field Crystal Models as a Lense on Nonequilibrium Kinetic Pathways

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# Abstract

Abstract goes here



# Dedication

To mum and dad





# Declaration

I declare that..



# Acknowledgements

I want to thank...



# Contents



## Chapter 1

# Introduction





## Chapter 2

# Microscopic Origins of Phase Field Crystal Theory

In a many body system in which particles interactions are independent of their velocities, we may split contributions to the free energy into two parts: the ideal and the excess.

### The ideal component of the free energy

The ideal component comes from the kinetic energy term in the Hamiltonian and is known exactly.

$$\beta\mathcal{F}_{id}[\rho] = \int dr \rho(r) \{ \ln( \Lambda^3 \rho(r) ) - 1 \} \quad (2.1)$$

Where,

$\Lambda^3$  is the thermal DeBroglie volume

$\rho(r)$  is the number density

$\beta$  is the inverse temperature ( $1/k_b T$ )

### Expansion of the excess free energy

The excess component comes from the interaction term in the Hamiltonian and is often not known exactly and must be modelled. A common technique for modelling the excess free energy is to expand it around uniform fluid reference state. This reference state is characterized by a number density,  $\rho_0$ , and chemical potential  $\mu_0$ .

$$\beta\mathcal{F}_{ex}[\rho] = \beta\mathcal{F}_{ex}^0 + \beta \int dr \left. \frac{\delta\mathcal{F}_{ex}}{\delta\rho} \right|_{\rho=\rho_0} \Delta\rho + \beta \frac{1}{2} \int dr \int dr' \Delta\rho(r') \left. \frac{\delta^2\mathcal{F}_{ex}}{\delta\rho(r)\delta\rho(r')} \right|_{\rho=\rho_0} \Delta\rho(r') + \dots \quad (2.2)$$

Where,

$\Delta\rho$  is difference from reference density ( $\rho(r) - \rho_0$ )

$\mathcal{F}_{ex}^0$  is the free energy of the reference state

The excess free energy is the generating function of direct correlation functions  $C^{(n)}(r_1, \dots, r_n)$ . In particular this means that direct correlation functions can be written as,

$$C^{(n)}(r_1, \dots, r_n) = -\beta \frac{\delta^{(n)} \mathcal{F}_{ex}[\rho]}{\delta \rho(r_1) \dots \delta \rho(r_n)}, \quad (2.3)$$

and our previous expansion can be rewritten in terms of these direct correlation functions.

$$\beta \mathcal{F}_{ex}[\rho] = \beta \mathcal{F}_{ex}^0 - \int dr C_0^{(1)}(r) \Delta \rho + \beta \frac{1}{2} \int dr \int dr' \Delta \rho(r') C_0^{(2)}(r, r') \Delta \rho(r') + \dots \quad (2.4)$$

In the absence of an external field the single particle direct correlation function of the reference system is simply  $\beta \mu_0^{ex}$ .

### Total Free Energy

If we now add in the ideal contribution to the free energy and take advantage of the fact that the excess chemical potential of the reference fluid can be written as the total chemical potential minus the ideal contribution,

$$\mu_0^{ex} = \mu_0 - \mu_0^{id} = \mu_0 - k_b T \ln(\Lambda^3 \rho_0). \quad (2.5)$$

to find an expression for the total free energy:

$$\beta \mathcal{F}[\rho] = \beta \mathcal{F}_0 + \int dr \left\{ \Delta \rho \ln \left( \frac{\Delta \rho}{\rho_0} \right) - (1 - \mu_0) \Delta \rho \right\} - \frac{1}{2} \int dr \int dr' \Delta \rho(r) C_0^{(2)}(r, r') \Delta \rho(r') \quad (2.6)$$

### Smooth atom approximation and the PFC Free Energy

To construct the phase-field crystal free energy we assume that the density fluctuations,  $\Delta \rho(r)$ , are small and expand the logarithm term to quartic order in the fluctuations. Furthermore, we nondimensionalize the free energy by scaling out the reference density,  $\rho_0$ , and changing variables to  $n(r) = \Delta \rho(r)/\rho_0$ .

$$\frac{\beta \mathcal{F}[n]}{\rho_0} = \frac{\beta \mathcal{F}_0}{\rho_0} + \int dr \mu_0 \rho_0 n(r) + \frac{n^2(r)}{2} - \frac{n^3(r)}{6} + \frac{n^4(r)}{12} - \frac{\rho_0}{2} \int dr \int dr' n(r) C_0^{(2)}(r, r') n(r') \quad (2.7)$$

At this point we note that the constant and linear terms in the free energy can be removed without changing the properties of the functional and we are left with the following minimal free energy functional:

$$\frac{\beta \mathcal{F}}{\rho_0} = \int dr \frac{n^2(r)}{2} - \frac{n^3(r)}{6} + \frac{n^4(r)}{12} - \frac{1}{2} \int dr \int dr' n(r) C^{(2)}(r, r') n(r'). \quad (2.8)$$

Note, that by convention a factor of the reference density is absorbed into the pair correlation function in the PFC free energy functional.

## Chapter 3

# Theory of Phase Field Crystal Models

### 3.1 Derivation of the General Binary XPFC Free Energy

Now that we've seen the recipe for building a phase-field crystal theory for a single component system the process for building a multicomponent theory proceeds analogously with a few minor changes in perspective. We start again by looking at the ideal and excess contributions to the free energy.

#### Ideal Free Energy to Two Components

The kinetic energy terms of each species in the Hamiltonian give rise to separate contributions to the free energy as you might expect. We'll label the two species A and B.

$$\beta\mathcal{F}_{id}^{tot}[\rho_A, \rho_B] = \beta\mathcal{F}_{id}[\rho_A] + \beta\mathcal{F}_{id}[\rho_B] \quad (3.1)$$

Where,

$\beta\mathcal{F}_{id}$  is the same ideal free energy functional as previously

#### Excess Free Energy of a Two Component System

Our expansion of the excess free energy works just as before but we must sum over the contributions from each species.

$$\beta\mathcal{F}_{ex} = \beta\mathcal{F}_{ex}^0 - \int dr C_i^{(1)}(r) \Delta\rho_i(r) - \frac{1}{2} \int dr \int dr' \Delta\rho_i(r) C_{ij}^{(2)}(r, r') \Delta\rho_j(r') \quad (3.2)$$

Where indices denote species (A or B) and repeated indices are summed over.

#### Total free energy of a Two Component System

Putting together the excess and ideal terms together and dropping the constant and linear terms as we did previously we find the following total free energy,

$$\beta\mathcal{F}[\rho_A, \rho_B] = \int dr \left\{ \Delta\rho_i \ln \left( \frac{\Delta\rho_i}{\rho_{i0}} \right) - \Delta\rho_i \right\} - \frac{1}{2} \int dr \int dr' \Delta\rho_i(r) C_{ij}^{(2)}(r, r') \Delta\rho_j(r') \quad (3.3)$$

**Changing variables**

Typically, the concentration is the variable we care about in binary systems so instead of preceeding with the usual phase field crystal approximations at this point we make a change of variables to concentration,  $c$ , and total density,  $\rho$ .

$$\begin{aligned}\rho &= \rho_A + \rho_B & \rho_0 &= \rho_{0A} + \rho_{0B} \\ c &= \frac{\rho_A}{\rho_A + \rho_B} & c_0 &= \frac{\rho_{0A}}{\rho_{0A} + \rho_{0B}}\end{aligned}$$

Making this change of variables and seperating total density and concentration contribution we find a free energy functional of the form,

$$\beta\mathcal{F}[c, \rho] = \tag{3.4}$$

Chapter 4

Chapter Three Title



Chapter 5

Chapter Four Title





## Chapter 6

# Conclusion



## Appendix A

# Noise in Nonlinear Langevin Equations

When using Langevin equations to study non-equilibrium statistical mechanics the noise strength can be linked to the transport coefficients through a generalization of the Einstein relation. The generalization was first developed by Onsager and Machlup [3]. The typical strategy for deriving such a relationship is to evaluate the equilibrium pair correlation function by two separate methods: the equilibrium partition functional and the equation of motion<sup>1</sup>.

While the equilibrium partition functional gives pair correlation through the typical statistical mechanical calculation, the equation of motion can be used to derive a dynamic pair correlation function that must be equal to the equilibrium pair correlation function in the long time limit.

In what follows we'll look at how to formulate a generalized Einstein relation from a generic Langevin equation and then calculate two specific examples using Model A dynamics and a  $\phi^4$  theory and Time Dependent Density Functional Theory (TDDFT) and a general Helmholtz free energy.

### A.1 Generalized Einstein Relations in an Arbitrary Model

We start by considering a set of microscopic observables,  $a_i(r, t)$ , that are governed by a nonlinear Langevin equation,

$$\frac{\partial \mathbf{a}(r, t)}{\partial t} = F[\mathbf{a}(r, t)] + \boldsymbol{\xi}(r, t). \quad (\text{A.1})$$

Where,  $\mathbf{a}$ , denotes a vector of our fields of interest. These microscopic equation of motion may have been derived from linear response, projection operators or some other non-equilibrium formalism. We assume that the random driving force,  $\boldsymbol{\xi}(r, t)$  is unbiased, Gaussian noise that is uncorrelated in time.

$$\langle \boldsymbol{\xi}(r, t) \rangle = 0 \quad (\text{A.2})$$

$$\langle \boldsymbol{\xi}(r, t) \boldsymbol{\xi}^\dagger(r', t') \rangle = \mathbf{L}(r, r') \delta(t - t') \quad (\text{A.3})$$

---

<sup>1</sup>For considerations far from equilibrium see [2, 4, 1]

We wish to constrain the form of the covariance matrix,  $\mathbf{L}$ , by demanding that the solution to the Langevin equation eventually decays to equilibrium and that correlations in equilibrium are given by Boltzmann statistics.

We begin by linearizing the equation of motion about an equilibrium solution,  $\mathbf{a}(r, t) = \mathbf{a}_{eq}(r) + \hat{\mathbf{a}}(r, t)$ .

$$\frac{\partial \hat{\mathbf{a}}(r, t)}{\partial t} = \mathbf{M}(r, r') * \hat{\mathbf{a}}(r', t) + \boldsymbol{\xi}(r, t) \quad (\text{A.4})$$

Where,  $*$  denotes an inner product and integration over the repeated variable. eg:

$$\mathbf{M}(r, r') * \hat{\mathbf{a}}(r') = \sum_j \int dr' M_{ij}(r, r') \hat{a}_j(r'). \quad (\text{A.5})$$

We can formally solve our linearized equation of motion,

$$\hat{\mathbf{a}}(r, t) = e^{\mathbf{M}(r, r')t} * \hat{\mathbf{a}}(r', 0) + \int_0^t d\tau e^{\mathbf{M}(r, r')(t-\tau)} * \boldsymbol{\xi}(r', \tau), \quad (\text{A.6})$$

And use this formal solution to evaluate the dynamic pair correlation function.

$$\begin{aligned} \langle \hat{\mathbf{a}}(r, t) \hat{\mathbf{a}}^\dagger(r', t') \rangle &= e^{\mathbf{M}(r, r_1)t} * \langle \hat{\mathbf{a}}(r_1, 0) \hat{\mathbf{a}}^\dagger(r_2, 0) \rangle * e^{\mathbf{M}^\dagger(r', r_2)t'} \\ &+ \int_0^t \int_0^{t'} d\tau d\tau' e^{\mathbf{M}(r, r_1)(t-\tau)} * \langle \boldsymbol{\xi}(r_1, 0) \boldsymbol{\xi}^\dagger(r_2, 0) \rangle * e^{\mathbf{M}^\dagger(r', r_2)(t'-\tau')} \end{aligned} \quad (\text{A.7})$$

To evaluate the equilibrium correlation function we take the limit as each time goes to infinity together ( $t = t' \rightarrow \infty$ ). It is important to note that every eigenvalue of  $\mathbf{M}$  must be negative for our solution to decay to equilibrium in the long time limit (eg.  $\lim_{t \rightarrow \infty} \hat{\mathbf{a}}(r, t) = 0$ ) and as such the first term in our dynamic correlation function won't contribute to the equilibrium pair correlation. This is as we might expect as the first term holds the contributions to the dynamic correlation function from the initial conditions. The second term can be evaluated by substituting the noise correlation and evaluating the delta function.

$$\boldsymbol{\Gamma}(r, r') = \lim_{t \rightarrow \infty} \langle \hat{\mathbf{a}}(r, t) \hat{\mathbf{a}}^\dagger(r', t) \rangle = \int_0^\infty dz e^{\mathbf{M}(r, r_1)z} * \mathbf{L}(r_1, r_2) * e^{\mathbf{M}^\dagger(r', r_2)z} \quad (\text{A.8})$$

Considering the product  $\mathbf{M}(r, r_1) * \boldsymbol{\Gamma}(r_1, r')$  and performing an integration by parts gives the final generalized Einstein relation.

$$\mathbf{M}(r, r_1) * \boldsymbol{\Gamma}(r_1, r') + \boldsymbol{\Gamma}(r, r_1) * \mathbf{M}^\dagger(r_1, r') = -\mathbf{L}(r, r') \quad (\text{A.9})$$

## A.2 Example 1 - Model A

As a first example of calculating an Einstein relation consider the following free energy functional under non-conservative, dissipative dynamics.

$$\beta \mathcal{F}[\phi] = \int dr \left\{ \frac{1}{2} |\nabla \phi(x)|^2 + \frac{r}{2} \phi^2(x) + \frac{u}{4!} \phi^4(x) + h(x) \phi(x) \right\} \quad (\text{A.10})$$

$$\frac{\partial \phi(x, t)}{\partial t} = -\Gamma \left( \frac{\delta \beta \mathcal{F}[\phi]}{\delta \phi(x)} \right) + \xi(x, t) \quad (\text{A.11})$$

The random driving force,  $\xi$ , is Gaussian noise, uncorrelated in time.

$$\langle \xi(x, t) \rangle = 0 \quad (\text{A.12})$$

$$\langle \xi(x, t) \xi(x', t') \rangle = L(x - x') \delta(t - t') \quad (\text{A.13})$$

To compute the Einstein relation for this theory we start by calculating the pair correlation function using the equilibrium partition function and Boltzmann statistics.

### A.2.1 The partition function route

In equilibrium the probability of particular field configuration is given by the Boltzmann distribution.

$$\mathcal{P}_{eq}[\phi] = \frac{e^{-\beta \mathcal{F}[\phi]}}{\mathcal{Z}[h(x)]} \quad (\text{A.14})$$

Where,  $\mathcal{Z}[h(x)]$  is the partition functional and is given by a path integral over all field configurations.

$$\mathcal{Z}[h(x)] = \int \mathcal{D}[\phi] e^{-\beta \mathcal{F}[\phi]} \quad (\text{A.15})$$

Evaluation of the partition function is of some importance because it plays the role of a moment generating function.

$$\frac{1}{\mathcal{Z}[h]} \frac{\delta^n \mathcal{Z}[h]}{\delta h(x_1) \dots \delta h(x_n)} = \langle \phi(x_1) \dots \phi(x_n) \rangle \quad (\text{A.16})$$

In general the partition function cannot be computed directly, but in the special case of Gaussian free energies it can. To that end we consider expanding  $\phi$  around an equilibrium solution,  $\phi(x) = \phi_0 + \Delta\phi(x)$ , and keeping terms to quadratic order in the free energy.

$$\beta \mathcal{F}[\Delta\phi] = \int dr \left\{ \frac{1}{2} \Delta\phi(x) \left( r - \nabla^2 + \frac{u}{2} \phi_0^2 \right) \Delta\phi(x) - h(x) \Delta\phi(x) \right\} \quad (\text{A.17})$$

Here the partition function is written in a suggestive form. As stated previously, functional integrals are difficult to compute in general, but Gaussian functional integrals do have a solution.

### Computing the Pair correlation function in the Gaussian approximation

To compute the pair correlation function we use the Fourier space variant of the partition function,

$$\mathcal{Z}[\tilde{h}(k)] \propto \exp \left\{ \frac{1}{2} \int dk \frac{h(k) h^*(k)}{r + \frac{u}{2} \phi_0^2 + |k|^2} \right\}. \quad (\text{A.18})$$

The pair correlation function,  $\langle \Delta\tilde{\phi}(k) \Delta\tilde{\phi}^*(k') \rangle$ , is then computed using equation A.16.

$$\langle \Delta\tilde{\phi}(k) \Delta\tilde{\phi}^*(k') \rangle = \frac{2\pi \delta(k + k')}{r + \frac{u}{2} \phi_0^2 + |k|^2} \quad (\text{A.19})$$

### A.2.2 The Equation of Motion Route

The equation of motion supplies a second method for evaluating the pair correlation function in equilibrium.

$$\frac{\partial \phi}{\partial t} = -\Gamma \left( (r - \nabla^2) \phi(x, t) + \frac{u}{3!} \phi^3(x, t) \right) + \xi(x, t), \quad (\text{A.20})$$

Our equation of motion, can be linearized around an equilibrium solution,  $\phi_0$ , just as we did in the partition function route to the pair correlation function. In a similar vain, we will Fourier transform the equation of motion as well.

$$\frac{\partial \Delta \tilde{\phi}(k, t)}{\partial t} = -\Gamma \left( \left( r + \frac{u}{2} \phi_0 + |k|^2 \right) \Delta \tilde{\phi}(k, t) \right) + \xi(k, t) \quad (\text{A.21})$$

Comparing with our generalized approach we can read of  $M(k, k')$  from the lineared equation of motion:

$$M(k, k') = -\Gamma \left( \left( r + \frac{u}{2} \phi_0 + |k|^2 \right) \delta(k + k') \right) \quad (\text{A.22})$$

Finally, once we compute the generalized Einstein relation with our specific pair correlation and  $M(k, k')$  we find,

$$L(k, k') = 2\Gamma \delta(k + k'), \quad (\text{A.23})$$

Or equivalently,

$$L(x, x') = 2\Gamma \delta(x - x'). \quad (\text{A.24})$$

## A.3 Example 2 - Time Dependent Density Functional Theory

In time dependent density functional theory (TDDFT) we have an equation of motion of the following form,

$$\frac{\partial \rho(r, t)}{\partial t} = D_0 \nabla \cdot \left[ \rho(r, t) \nabla \left( \frac{\delta \mathcal{F}[\rho]}{\delta \rho} \right) \right] + \xi(r, t) \quad (\text{A.25})$$

Where,  $D_0$  is the equilibrium diffusion constant and  $\xi$  is the stochastic driving force. We assume once again that the driving force has no bias, but we now allow the noise strength to be a generic kernel  $L(r, r')$ .

$$\langle \xi(r, t) \rangle = 0 \quad (\text{A.26})$$

$$\langle \xi(r, t) \xi(r', t') \rangle = L(r, r') \delta(t - t') \quad (\text{A.27})$$

### A.3.1 Pair Correlation from the Partition Functional

Just like with the  $\phi^4$  model we want to expand our free energy functional around an equilibrium solution. In this case our free energy functional is generic so this expansion is purely formal.

$$\mathcal{F}[\rho] = \mathcal{F}_{eq} + \beta \int dr \left( \frac{\delta \mathcal{F}[\rho]}{\delta \rho(r)} \right) \Big|_{\rho_{eq}} \Delta \rho(r) + \frac{1}{2} \int dr \int dr' \Delta \rho(r) \left( \frac{\delta^2 \mathcal{F}[\rho]}{\delta \rho(r) \delta \rho(r')} \right) \Big|_{\rho_{eq}} \Delta \rho(r') \quad (\text{A.28})$$

The first term we can neglect as it adds an overall scale to the partition function that will not affect any of moments. Second moment only shifts the average so we can ignore it as well and so we're left with a simple quadratic free energy once again.

$$\mathcal{F}[\rho] = \frac{1}{2} \int dr \int dr' \Delta\rho(r) \Gamma^{-1}(r, r') \Delta\rho(r') \quad (\text{A.29})$$

Where,  $\Gamma^{-1}(r, r')$  is the second functional derivative of the free energy functional in equilibrium. Computing the pair correlation function from the partition function yields, as might be expected,

$$\langle \Delta\rho(r) \Delta\rho(r') \rangle = \Gamma(r, r') \quad (\text{A.30})$$

### A.3.2 Linearizing the equation of motion

Linearizing the equation of motion about an equilibrium solution we find the following form,

$$\frac{\partial \Delta\rho(r, t)}{\partial t} = D_0 \nabla \cdot [\rho_{eq}(r) \nabla (\Gamma^{-1}(r, r') * \Delta\rho(r', t))] + \xi(r, t) \quad (\text{A.31})$$

Once again we can read of the kernel  $M(r, r')$  from the linearized equation.

$$M(r, r') = D_0 \nabla \cdot [\rho_{eq}(r) \nabla (\Gamma^{-1}(r, r'))] \quad (\text{A.32})$$

Plugging into the generalized Einstein relation, we find a the factors of the pair correlation cancel giving a simple form for the kernel  $L(r, r')$ .

$$L(r, r') = -2D_0 \nabla \cdot (\rho_{eq}(r) \nabla) \delta(r - r') \quad (\text{A.33})$$





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## Appendix B

# Gaussian Functional Integrals

### Gaussian Functional Integrals

In the study of the statistical physics of fields we often encounter functional integrals of the form,

$$\mathcal{Z}[h(x)] = \int \mathcal{D}[\phi] \exp \left\{ - \int dx \int dx' \left[ \frac{1}{2} \phi(x) \mathbf{K}(x, x') \phi(x') \right] + \int dx [h(x) \phi(x)] \right\}. \quad (\text{B.1})$$

Solutions to this integral are not only important in their own right but are also the basis of perturbative techniques. The detail of how to solve this integral can be found in [1] and are repeated here for the convenience of the reader.

This integral is simply the continuum limit of a multivariable Gaussian integral,

$$\mathcal{Z}[\mathbf{h}] = \int \prod_i dx_i \exp \left\{ - \frac{1}{2} \sum_i \sum_j x_i \mathbf{K}_{ij} x_j + \sum_i h_i x_i \right\}, \quad (\text{B.2})$$

For which the solution is,

$$\mathcal{Z}[\mathbf{h}] = \sqrt{\frac{2\pi}{\det(\mathbf{K})}} \exp \left\{ \frac{1}{2} \sum_i \sum_j h_i \mathbf{K}_{ij}^{-1} h_j \right\}. \quad (\text{B.3})$$

In the continuum limit, the solution has an analogous form.

$$\mathcal{Z}[h(x)] \propto \exp \left\{ \int dx \int dx' \left[ \frac{1}{2} h(x) \mathbf{K}^{-1}(x, x') h(x') \right] \right\} \quad (\text{B.4})$$

Where  $\mathbf{K}^{-1}$  is defined by,

$$\int dx' \mathbf{K}(x, x') \mathbf{K}^{-1}(x', x'') = \delta(x - x''). \quad (\text{B.5})$$

Ultimately, we don't need to worry about the constant of proportionality in equation B.4 because we'll be dividing this contribution when calculating correlation functions.



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