

Numerical Simulation and Role of Noise in the Cahn-Hilliard-Cook Equation below the Critical Dimension

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ABSTRACT

Introducing stochastic or thermal noise into partial differential equations that would otherwise be deterministic is a useful technique for the analysis of ensemble simulations. It can also aid the unfreezing of structural growth barriers that occur in spatial growth systems. This article presents some modelling experiments using Cook's noise term in the Cahn-Hilliard-Cook equation simulated on multi-dimensional spatial meshes. We show that below the critical dimension noise is actually necessary to enable long-term spatial growth behaviours that are sufficiently driven by surface tension effects in higher dimensions. We also discuss numerical methods for the study using finite differencing techniques.

KEY WORDS

numerical methods; simulation; model development; statistical modelling; Cahn Hilliard Cook equation; stochastic noise.

1 Introduction

The Cahn-Hilliard (CH) equation [1] models phase separation in a material system such as an alloy. It has recently attracted new attention as it can also be used to model the dynamics of related to fluid separation dynamics under certain conditions. Simulated as a partial differential equation (PDE), the CH model system exhibits initial domain growth from a quenched random mixture to the spinodal domain coarsening time regime. While early growth stages agree with experimental exponential growth laws, in the long term the model system can become "stuck" and the domains become essentially frozen. Experimental data suggests that real systems continue to develop and coarsen and so the freezing of a model system may be attributable to finite size effects or to unrealistic thermal coupling.

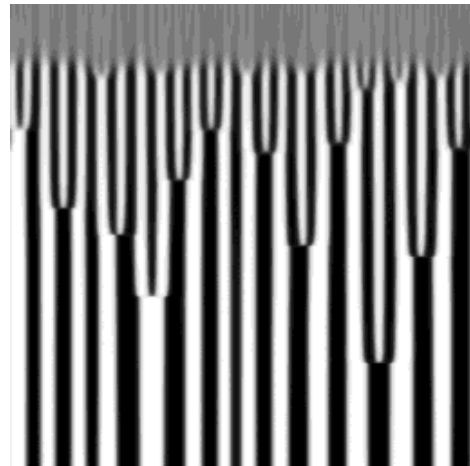


Figure 1: Domain formation and coarsening in the Cahn-Hilliard equation in 1-D (1-d space is across the page and time is shown down the page.)

Figure 1 shows the time evolution of a one dimensional Cahn-Hilliard model system where the initial system is quenched from a random black/white mixture and domains of black or white material form and coarsen. The model system is shown to become stuck and the spatial domains stop coarsening. The Cahn-Hilliard equation and model both have a relatively long history [2] Considerable work has been reported in the literature on numerical work simulating the CH equation, particularly in two dimensional systems. Some theoretical work is still being done on the CH model [3–5] but most reported work makes use of numerical methods.

Generally the equation can be solved using a number of different numerical approaches. Finite difference methods [6] are still widely used for solving CH system on regular meshes [7], although finite element [8], multi-grid

methods [9] and spectral methods [10] have also become popular relatively recently. In all cases, considerable numerical computation is required to adequately explore even medium sized meshes ($N > 100 \times 100$) for numerical integration simulation times long enough for spinodal and surface tension driven effects to take the system configuration to completion in terms of the spatial structures that are formed. It is possible to simulate small CH systems using problem solving software packages such as Matlab [11] but for reasons of computational speed and performance, most authors report using specially written numerical simulation codes – often using parallel computing techniques and libraries. Recent work using data parallel capabilities on general purpose Graphical Processing Units (GPUs) has opened up new possibilities for semi-interactive simulation [12] and visualisation [13] of the CH equation and PDE-based model systems like it.

There has been successful numerical work reported on the formation of transition layers [14] nonlinear transitions between domain nucleation and spinodal [15] in the CH system, as well as on domain wall formation [16], non local effects [17] and viscosity effects [18]. Fewer numerical studies appear to have been done on modelling the full Cahn-Hilliard-Cook (CHC) model [19–21] Some work is reported on the use of stochastic Monte Carlo approaches [22] and Davydov *et al.* report work on freezing-in structural effects [23].

The Cook noise term [24] is introduced in an attempt to model thermal effects into the equation so that a source of fluctuations is available to assist in unfreezing the long term behaviour. This noise term has been studied at a theoretical level but surprisingly little work has been done on long term simulations of large model system simulations.

In this paper we explore the quantitative effects of various magnitudes of the Cook noise term on simulations of the full Cahn-Hilliard-Cook (CHC) partial differential equation. We explore the effect of the stochastic term on what is otherwise a deterministic field equation and its effect in overcoming barriers to domain coarsening and growth. A number of properties of the model system can be studied including the characteristic length scales of domains and the field susceptibility. A useful metric for detecting slowing-down effects in domain-coarsening however is a count of the number of spatial domains present in the model system with time. This provides a quantitative supplement to the qualitative visual evidence for slowing down that can be obtained from graphic visualisation of the model field in one, two or three dimensions [25].

We present a brief derivation of the relevant aspects of the Cahn-Hilliard equation in section 2 as well as a review of how the Cook noise term is introduced theoretically in section 3 and practically as part of a simulation in section 4 using numerical methods (section 5). In section 6 we present some results from various simulations and offer some discussion of the unfreezing effects of the noise

term in section 7 as well as some concluding remarks and ideas for further work on applications with other stochastic equations.

2 Cahn-Hilliard-Cook Equation

In this section we give a brief derivation of the Cahn-Hilliard equation based on the treatment given in [26] before reviewing the effect of Cook’s noise term that extends it to a stochastic model system in section 4.

A useful starting point is to consider a binary alloy comprising atomic species A and B and a continuous concentration field variable that gives the excess density of species A over B. The Cahn-Hilliard theory is based on a smoothly varying field variable $\phi(\mathbf{r})$ that represents the average spatial concentration over a unit cell. We employ the field variable in units so that it lies on the interval $[-1, 1]$ with $+1$ representing A species atoms and -1 representing B-species atoms. For illustrative purposes, we colour these field domains either black and white or with a false colour representation in the figures shown in this paper.

We can derive the Cahn-Hilliard-Cook equation starting from the Helmholtz free energy expression for a binary mixture with inhomogenous composition and the Ginzberg-Landau energy functional [27] can be used for this.

$$\frac{\mathcal{F}\{\phi(\mathbf{r})\}}{k_b T} = \frac{F_0}{k_b T} + \int_V \left\{ f\{\phi(\mathbf{r})\} - \frac{H}{k_b T} \phi(\mathbf{r}) + \frac{1}{2d} [R \nabla \phi(\mathbf{r})]^2 \right\} d\mathbf{r} \quad (1)$$

The scalar field $\phi(\mathbf{r})$ describes the atomic composition as a function of spatial position \mathbf{r} where the range of interaction is given by $\frac{R}{d}$ and the applied field is H . In the case of a fixed atomic concentration ratio, we set the applied field H to zero to model an equal concentration of A-type and B-type atoms.

The phase-separating alloy may be modelled as a set of macroscopic cells, each containing a volume of space and a number of atomic sites. These cells must be large enough for a local instantaneous free energy function $f(\mathbf{r})$ to be defined, but small enough that the effect of ‘relevant’ short length scale composition fluctuations are not integrated out. It is convenient to write this We employ the Landau form of the localised free energy density function $f\{\phi(\mathbf{r}, t)\}$:

$$f\{\phi(\mathbf{r}, t)\} = f_0 - \frac{1}{2} b (\phi(\mathbf{r}, t))^2 + \frac{1}{4} u (\phi(\mathbf{r}, t))^4 + \dots \quad (2)$$

where $b, u > 0$. We express conservation of atoms of a particular species using the constraint:

$$\frac{1}{V} \int_V \phi(\mathbf{r}, t) d\mathbf{r} = c_A \quad (3)$$

where V gives the system's volume, and we denote the concentration of atomic species A by c_A . As a consequence of this obtain we obtain a continuity law:

$$\frac{d\phi(\mathbf{r}, t)}{dt} + \nabla \cdot \mathbf{j}(\mathbf{r}, t) = 0 \quad (4)$$

This concentration current $\mathbf{j}(\mathbf{r}, t)$ is taken to be proportional to the gradient of the *localised* chemical potential difference $\mu(\mathbf{r}, t)$ with a constant of proportionality m known as the mobility.

$$\mathbf{j}(\mathbf{r}, t) = -m\nabla\mu(\mathbf{r}, t) \quad (5)$$

The definition of the chemical potential difference is:

$$\mu(\mathbf{r}, t) = \frac{d\mathcal{F}\{\phi(\mathbf{r}, t)\}}{d\phi(\mathbf{r}, t)} \quad (6)$$

with the Landau functional \mathcal{F} given above in equation 1. When we differentiate this functional with respect to ϕ and take the mobility to be a simple scalar we obtain the chemical potential difference:

$$\mu(\mathbf{r}, t) = \left. \frac{\partial f}{\partial \phi} \right|_T - \frac{R^2}{d} k_b T \nabla^2 \phi(\mathbf{r}, t) \quad (7)$$

This is substituted into the continuity equation 4 to yield the Cahn-Hilliard equation [27] in terms of the concentration field variable.

$$\frac{\partial \phi(\mathbf{r}, t)}{\partial t} = m \nabla^2 \left(\left. \frac{\partial f(\phi(\mathbf{r}, t))}{\partial \phi} \right|_T - K \nabla^2 \phi(\mathbf{r}, t) \right) \quad (8)$$

The parameter K is defined as:

$$K = \frac{R^2}{d} k_b T \quad (9)$$

Expanding equation 8 we obtain:

$$\frac{\partial \phi}{\partial t} = m \nabla^2 (-b\phi + u\phi^3 - K \nabla^2 \phi) \quad (10)$$

Where we follow usual practice and truncate the power series in the free energy at the fourth-order ϕ term [28].

3 Cahn-Hilliard Theory

Equation 8 cannot be solved in closed form since it is non-linear. Numerical methods are therefore necessary. Cahn-Hilliard theory [27] involves linearising the equation around the specific point $\phi = c_A$ from which we can obtain:

$$\frac{\partial(\phi - c_A)}{\partial t} = m \nabla^2 \left(\left. \frac{\partial f}{\partial \phi} \right|_{T, c_A} - K \nabla^2 (\phi - c_A) \right) \quad (11)$$

This can be Fourier-transformed with:

$$\Phi(\mathbf{q}, t) = \int e^{i\mathbf{q} \cdot \mathbf{r}} (\phi(\mathbf{r}, t) - c_A) d\mathbf{r} \quad (12)$$

from which we obtain:

$$\Phi(\mathbf{q}, t) = \Phi(\mathbf{q}, t=0) e^{B(\mathbf{q})t} \quad (13)$$

where the amplification factor B in time is:

$$B(\mathbf{q}) = -m\mathbf{q}^2 \left(\left. \frac{\partial f}{\partial \phi} \right|_{T, c_A} + K\mathbf{q}^2 \right) \quad (14)$$

The standard structure function can be then obtained as an auto-correlation of the concentration field variable:

$$S(\mathbf{q}, t) = \langle \Phi(-\mathbf{q}, t) \Phi(\mathbf{q}, t) \rangle|_T \quad (15)$$

It is useful to re-express this in terms of the values in effect just prior to the quench:

$$S(\mathbf{q}, t) = \langle \Phi(-\mathbf{q}, t=0) \Phi(\mathbf{q}, t=0) \rangle|_T e^{2B(\mathbf{q})t} \quad (16)$$

where prefactor term in equation 16 is then the static structure factor of the initial state at T_0 , just prior to the thermal quench.

$$S_0(\mathbf{q}) = \langle \Phi(-\mathbf{q}, t=0) \Phi(\mathbf{q}, t=0) \rangle|_{T=T_0} \quad (17)$$

The linearised Cahn-Hilliard theory indicates that fluctuations of \mathbf{q} that are present in the initial state will consequently grow exponentially with time after the thermal quench if $B(\mathbf{q}) > 0$ but would decay to zero for the case of $B(\mathbf{q}) < 0$. This gives a critical wave-vector \mathbf{q}_c for which the condition $B(\mathbf{q}) \equiv 0$ holds. Although there has been disagreement in the literature about the validity of the Cahn-Hilliard theory [29] it is known to be **invalid** for short range alloy type interactions and therefore full numerical methods are necessary to perform the simulations described in our model.

4 Cook's Noise Term in Cahn-Hilliard

The Cahn-Hilliard theory is a mean-field theory. It ignores thermal fluctuations and approximates the interactions between the concentration variables by a mean value. It is surprising how well it describes a sensible dynamic behaviour of a real system. Cook [24] gave an extension to the Cahn-Hilliard model based upon a random force term ζ .

$$\frac{\partial \phi(\mathbf{r}, t)}{\partial t} = m \nabla^2 \left(\left. \frac{\partial f(\phi(\mathbf{r}, t))}{\partial \phi} \right|_T - K \nabla^2 \phi(\mathbf{r}, t) \right) + \zeta(\mathbf{r}, t) \quad (18)$$

The very short time scale phonon modes of the alloy are conjectured to give rise to this noise. The ζ term has to have some specific properties:

$$\langle \zeta(\mathbf{r}, t) \rangle = 0 \quad (19)$$

$$\langle \zeta(\mathbf{r}, t) \zeta(\mathbf{r}', t') \rangle = -2k_b T m \nabla^2 \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \quad (20)$$

The first of these equations(19) mandates that there is no overall drift force and the second (20) that while the noise is uncorrelated in time, it is partially correlated in space. This is necessary so that the applied term is free from any long wavelength components in the noise spectrum.

The noise magnitude is adjusted by $k_b T$ and also the mobility m . The Laplacian term ∇^2 in equation 20 expresses the field-conservation law. In effect this means that a randomly applied force that adds atomic matter at one spatial site must be precisely balanced by force contributions at neighbouring sites which remove from those sites the exact same total amount of matter.

Addition of the Cook noise term is insufficient to rescue the linearised Cahn-Hilliard theory from invalidity. It does appear to have a useful tole to play in numerical simulations. It has been controversial whether the Cook noise term plays an important role in the long-term dynamics of the alloy [6]. The work we report on in this paper uses the Cook noise term in simulations to allow the phase-separating field to unfreeze when it becomes stuck in a one dimensional system.

5 Numerical Methods

In this work the CH and CHC equations are solved using finite-difference methods on regular hyper-cubic meshes in 1-D, 2-D and 3-D. For illustrative purposes the code below shows how the right hand side of the CH equation is computed for the k' th grid point and can be used in a midpoint or Runge-Kutta integration formula to update the grid of field values at each time step.

Listing 1: Stencil program source code (in C) for the right hand side (RHS) of the Cahn-Hilliard-Cook equation in 1-D.

```

rhs [k] = dt *
(
  - b *
    ( phi_zyxm1 + phi_zyxp1 -2.0 * phi_zyx )
  + U *
    ( pow(phi_zyxm1,3) +
      pow(phi_zyxp1,3) - 2.0 * pow(phi_zyx,3)
    )
  - Kbydx * ( phi_zyxm2 + phi_zyxp2
  - 4.0 * ( phi_zyxm1 + phi_zyxp1 )
  + 6.0 * phi_zyx )
  + zeta * noise
);

```

Note that the CH equation 8 involves a double Laplacian operator, which is implemented as a doubly applied stencil based on the gathered spatial variables around mesh point x, y, z at $x \pm 1, y \pm 1, z \pm 1$ which we denote in the program source as `xp1, xm1` etc. Similar formulations are

possible (but too lengthy to reproduce here) for 2-D and 3-D model meshes.

For the work reported in this paper we used a second-order time integration and a second-order spatial stencil method based on centred differences as shown. For high noise values a fourth-order time integration method was used to ensure the equation did not become numerically unstable. The work reported used a time step of $dt \equiv 0.01$ or 0.025 throughout. All our work reported here used custom C/C++ program source code in the style shown above.

The CH equation is recovered from code for the CHC by setting the ζ noise term prefactor to zero. The actual noise field addition can be generated using a number of algorithms. The simplest (global algorithm) is to create a floating point variable field array with the same dimensions as the model field ϕ and to add a distribution of random numbers to it. These can be either distributed uniformly or as a normal distribution generated from uniform deviates by – for example – a Box-Muller transformation [30]. In either case the noise constraints are satisfied by subtracting the mean value of the whole noise array from each cell value to ensure there is no overall drift applied.

Another technique is to iteratively apply a small random perturbation at each spatial site and immediately calculate the cumulative amount by which its 2, 4 or 6 neighbouring sites, in 1-, 2- or 3-dimensions respectively, would need to be adjusted to ensure locality of conservation. This spatially local algorithm is more expensive to compute, since it involves a manipulation of nearest neighbour terms. In the work reported here no difference was discerned between these two algorithms nor between application of uniform or Gaussian normally-distributed noise.

6 Numerical Results

We present a number of simulated model configuration snapshots and some measured properties showing how the CH and CHC equations behave in the long time limit. In each case model systems were initialised with a spatial ϕ field randomly distributed on $[-1, 1]$ and time integrated, deterministically for the CH equation or with the addition of stochastic noise in the case of CHC model systems. Configuration snapshots were visualised using customised OpenGL graphics software.

Figure 2 shows the time evolution of the CH model on a $d = 3, N = 32 \times 32 \times 32$ cubic mesh time integrated for up to time $t = 1024 = 102,400 \times 0.01$. A rainbow colour scheme shows the field variable ϕ from -1 (red) through zero (green) to $+1$ (blue). The system eventually reaches its long term stable limit of exactly two interleaved domains, initiated as many random fluctuations, then evolved by surface tension effects and shown wrapped around by the periodic boundary conditions. All other fluctuations and small initial domains have “evaporated” or been drawn

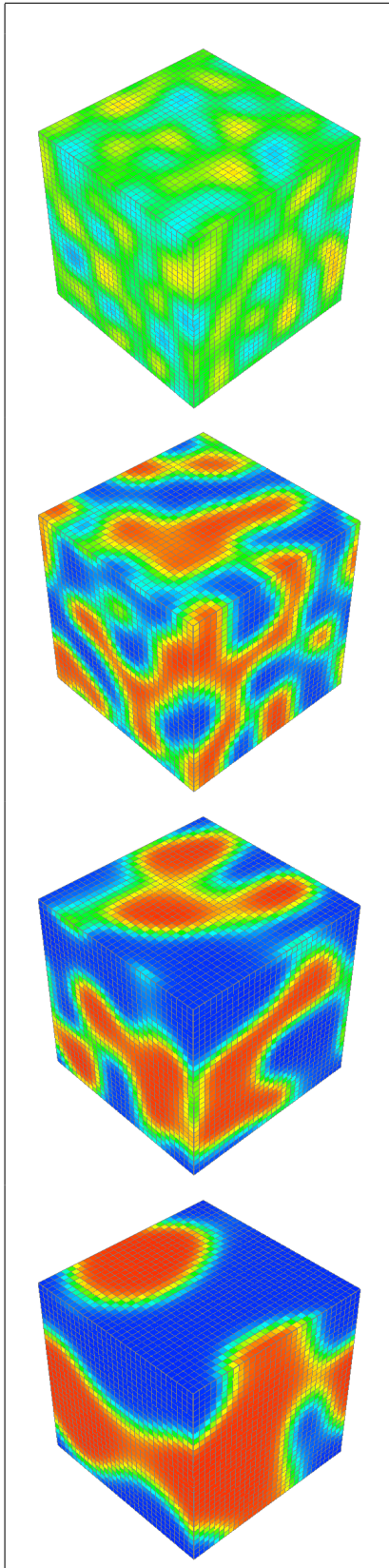


Figure 2: \log_2 Time Evolution of a $32 \times 32 \times 32$ CHC System at times: 0; 32; 128; 1024.

into the two main domains. Addition of Cook noise to the 3-D model system as shown (and 2-D models, not shown) made no material difference. The system has enough symmetry above the critical dimension of $d = 2$ that the Cook noise is not necessary for it to evolve to a long term stable spatial structure.

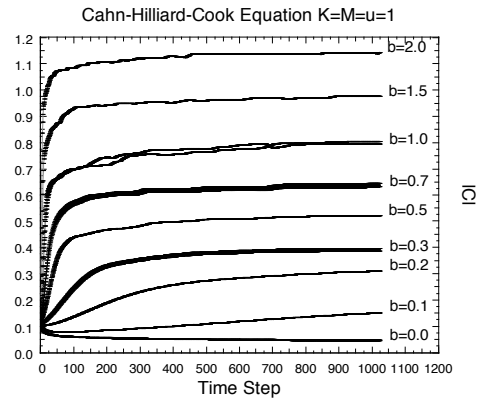


Figure 3: Mean concentration variation with inverse temperature b in the CHC Equation in 1D

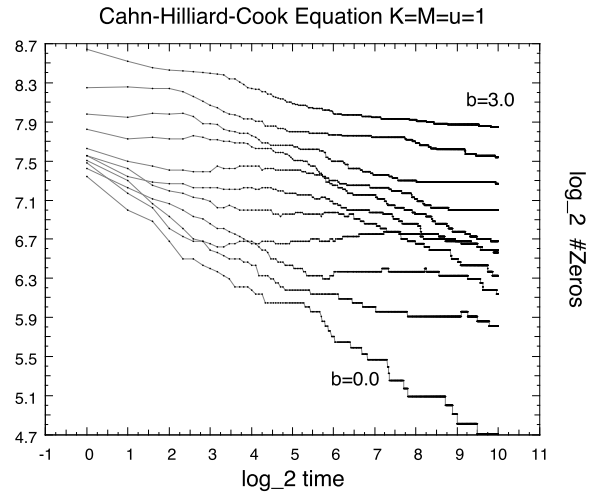


Figure 4: Number of Zeros (domains) in the CHC Equation

Figure 3 shows how the mean concentration (the average of the absolute value of the field ϕ) evolves with integration time. It always converges, albeit slowly and logarithmically, to a long term value, determined by the inverse temperature parameter b . The only exception being when $b = 0$ and the temperature is effectively infinite, the system is unable to sustain spatial domains and the mean

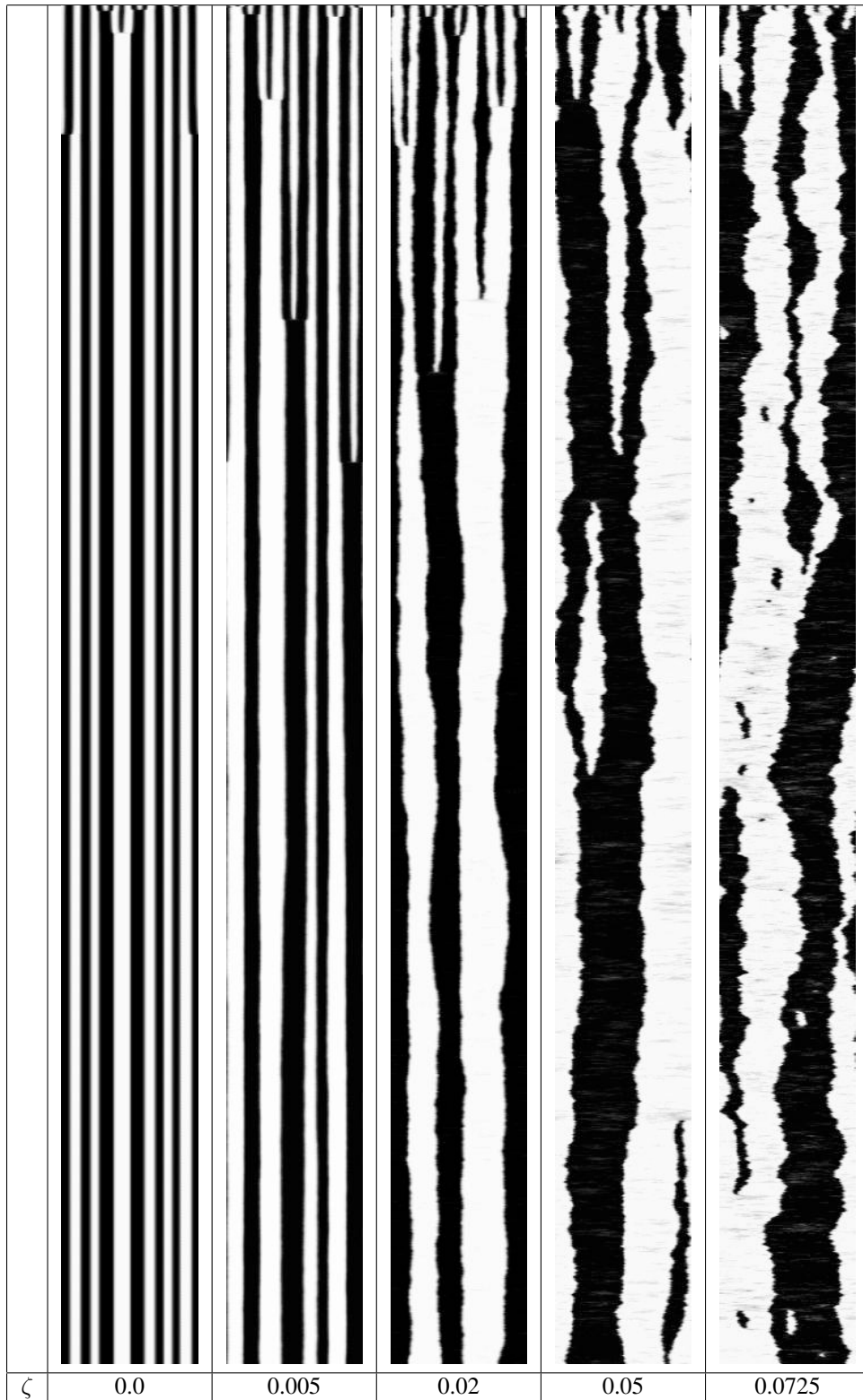


Figure 6: Effect of the Cook Noise term ζ on the CHC Equation in 1D. 0.05 is sufficient to reach just two (periodically connected) domains.

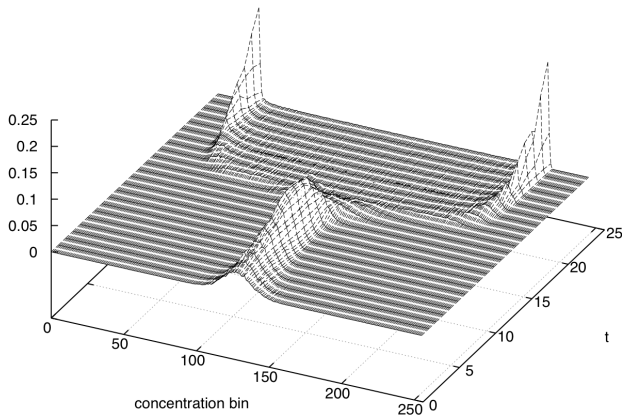


Figure 5: Concentration evolution in 100x100 system logarithmic time in units of 10^5 (4,000,000 by 0.025), $b=u=1$, noise=0

absolute final value of the field tends to zero. The critical temperature effect is seen between $b = 0.1$ and $b = 0.2$ where the characteristic shape of the curve changes.

It is possible to automatically identify the number and size of domains present in the model system using a component labelling algorithm [31]. This approach is computationally expensive but works in all spatial dimensions. A simpler approach can be taken in 1-D systems where the number of domains is related to the number of zeros in the field variable $\phi(x)$. Figure 4 shows a plot of the number of zeros and is thus an indication of how the number of spatial domains drops with time as the spinodal decomposition and surface driven effects combine the domains into one another. The data shown is quite noisy and would need to be averaged over a very large number of independent system runs to become less noisy. However it is indicative of the domain barriers.

Another useful indicator of the system behaviour is the histogram of ϕ field values present in the spatial system. This is shown histogrammed into 256 bins in figure 5 representing values from $\phi = -1$ through zero up to $\phi = +1$. The surface plot shows how the system is initially configured at time zero with a normal distribution of ϕ values and how this gradually phase separates out into the two peaks. the time scale shown is logarithmic. The two peaks correspond to black or white phases of figure 1 or the red and blue phases (using a colour rainbow palette) of figure 2. The data shown is for a 2-D system but again it is unaffected by the addition of Cook noise.

Figure 1 shows the characteristic phase separation behaviour in a 1-D CH model system. The initial small fluctuations do grow into large domains but in 1-D they become frozen, and even if the system is time-integrated for a very long time the domains are unable to combine together and remain fixed at relatively small length scales.

Figure 6 shows the effect of adding noise to a 1-D system for various values of the Cook noise strength param-

eter ζ . Sufficient noise helps the frozen in domains “find one another” and the spinodal decomposition can continue to completion and reach a state of exactly two domains present – one of each phase. Without noise, the system remains frozen or locked into multiple small-sized and isolated domains.

7 Discussion and Conclusions

We have presented simulations of the Cahn-Hilliard and Cahn-Hilliard-Cook models in multiple dimensional models simulated on regular meshes using finite difference methods. The “freezing-in” of domains that occurs in one dimension can be avoided if sufficient Cook noise is added into the model field as the CH equation is integrated. This is explained by the critical dimensionality of the system. It exhibits phase transitional behaviour and for dimensions at and above the critical dimension of $d = 2$ the noise term is not necessary. Rather, the initial randomness introduced when the field was initialised at time zero is sufficient for domains to nucleate and grow to completion in finite time.

The model also seems to be insensitive to the exact nature of the Cook noise term employed. One dimensional unfreezing worked equally well with either uniform or Gaussian noise. No discernable effect was observed in two or three dimensions regardless of the sort of noise term used. Similarly no effect was observed on using a global or local generated noise field.

The numerical methods described were adequate to simulate quite large model systems and for long integration times even with the addition of the destabilising noise term. The finite difference approach is not necessarily the most computationally efficient for this system but it is the most easily understood and analysed when new terms are being added experimentally. The C/C++ source code was adequate for the present study but data parallel techniques are important if even larger system sizes and even more experimental runs are needed in a study.

For areas of future study, there are a number of other effects in Cahn-Hilliard model systems such as striated string-like structural growth [32] and variable mobility [33] that may also be affected by introduction of a Cook noise term. This approach of adding a stochastic term to an otherwise deterministic partial differential equation find other uses in ensemble simulation but also for other PDE based system such as financial modelling [34].

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