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Perturbation Solution of the Cahn Hilliard Equations

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Abstract— The Spinodal Decomposition equations, as proposed by Cahn and Hilliard, are described by a fourth-order partial differential equation (PDE) that remains unsolved analytically up to the present time. This article delves into an exploration of these equations in their various forms, investigating solutions for both steady-state and transient scenarios using a variety of methodologies. Furthermore, the study addresses the moving boundary associated with the transformation through the lens of heat transfer principles. An attempt is made to solve this boundary using a combination of series and similarity transformation approaches.

The equation under consideration is believed to consist of two components: one stemming from the conventional Laplacian and Fickian contributions, for which Stefan solutions are well-established, and another involving a Double Laplacian term that has yet to be thoroughly investigated. To facilitate the analysis, the thin film approximation is employed to simplify certain aspects and to facilitate the derivation of a numerical solution

Introduction

The Cahn-Hilliard equation describes the process of phase separation in binary mixtures, alloys and polymer blends. This study focuses on the perturbative solutions of the one-dimensional Cahn-Hilliard equation with a double-well free energy potential, specifically applied to thin films.

Cahn-Hilliard Equation with Double-Well Potential. The Cahn-Hilliard equation in one dimension with a constant mobility \(M\) and a double-well free energy potential is given by:

$$\frac{\partial u}{\partial t} = M\nabla \cdot \left[\epsilon^2 \nabla^2 u - f'(u)\right] \quad (1)$$

where u is the order parameter,epsilon is a parameter related to the interfacial width, and f(u) is the free energy density with a double-well form given by

$$f(u) = \frac{1}{4}u^4 - \frac{1}{2}u^2 \quad (2)$$

Linearization and Fourier Transform. Starting with a uniform state and introducing a small perturbation, let:

$$u(\xi, \tau) = u_0 + u_1(\xi, \tau) \quad (3)$$

Assuming $u_0 = 0$ (a solution of the unperturbed system), the equation becomes:

$$\frac{\partial u_1}{\partial \tau} = -M\nabla^2\left(\epsilon^2\nabla^2 u_1 + u_1\right) \tag{4}$$

Applying the Fourier transform:

$$\hat{u}_1(k,\tau) = \hat{u}_1(k,0)\exp\left(-Mk^2(\epsilon^2k^2+1)\tau\right) \tag{5}$$

Thin Films and Fourier Series Expansion. For thin films with no-flux boundary conditions at$\xi = 0$ and $\xi = L$, we use a cosine series expansion:

$$u_1(\xi,\tau) = \sum_{n=0}^{\infty} a_n(\tau)\cos\left(\frac{n\pi\xi}{L}\right) \tag{6}$$

Substituting this into the linearized equation, we obtain:

$$\frac{da_n}{d\tau} = -M\left(\epsilon^2\left(\frac{n\pi}{L}\right)^4 + \left(\frac{n\pi}{L}\right)^2\right)a_n \tag{7}$$

Solving the ODE for the coefficients:

$$a_n(\tau) = a_n(0)\exp\left(-M\left(\epsilon^2\left(\frac{n\pi}{L}\right)^4 + \left(\frac{n\pi}{L}\right)^2\right)\tau\right) \tag{8}$$

The final solution for u_1 in a thin film is:

$$u_1(\xi,\tau) = \sum_{n=0}^{\infty} a_n(0)\cos\left(\frac{n\pi\xi}{L}\right)\exp\left(-M\left(\epsilon^2\left(\frac{n\pi}{L}\right)^4 + \left(\frac{n\pi}{L}\right)^2\right)\tau\right) \tag{9}$$

Evaluation of the First Few Terms. For very small ε , the term $\epsilon^2\left(\frac{n\pi}{L}\right)^4$ becomes negligible compared to $\left(\frac{n\pi}{L}\right)^2$. The first few terms are:

$$u_1(\xi,\tau) \approx \sum_{n=0}^{\infty} a_n(0)\cos\left(\frac{n\pi\xi}{L}\right)\exp\left(-M\left(\frac{n\pi}{L}\right)^2\tau\right) \tag{10}$$

Since $u(\xi,0) = u_0 + u_1(\xi,0) = u_0 + a_0(0) + a_1(0)\cos\left(\frac{\pi\xi}{L}\right)$, with boundary conditions

U(0) =a and u(L,) =b, we get:

From (3) a= u0 +u1(0,t) = u0 + $a_o(0)$ + $a_1(0)$
b= u0+ $a_0(0)$-$a_1(0)$ (11)
Since u0=0 (by condition)

$$u(\xi,\tau) = \frac{a+b}{2} + \frac{a-b}{2}\cos\left(\frac{\pi\xi}{L}\right)\exp\left(-M\left(\frac{\pi}{L}\right)^2\tau\right) \tag{12}$$

For smallξ, the approximation gives:

$$u_1 \approx a_0(0) + a_1(0)\left(1 - M\left(\frac{\pi}{L}\right)^2\tau\right)$$

~ (a+b)/2 +(a-b)/2 (1-M(/L)^2) (13)

Examining the growth of the two regimes (small time and small distance), take the ratio of the quantities:

$$\frac{1-\left(\frac{\pi\xi}{L}\right)^2/2}{1-M\left(\frac{\pi}{L}\right)^2\tau} \tag{14}$$

If L is large compared to ξ and $M\tau$, the expression can be expanded by the binomial theorem:

$$\left(1-\left(\frac{\pi\xi}{L}\right)^2/2\right)\left(1+M\left(\frac{\pi}{L}\right)^2\tau\right) \approx 1+M\left(\frac{\pi}{L}\right)^2\tau \tag{15}$$

Hence, the ratio grows linearly with time with the gradient $1+M\left(\frac{\pi}{L}\right)^2\tau$

Boundary Layer Thickness. Given the function $y=1-\frac{(\pi\xi)^2}{2L}$, we need to estimate the thickness of the boundary layer where the variable u changes rapidly. Using the definition of boundary layer as that where the variable drops to 1/e of its original value:

$$\delta=\sqrt{\frac{2L\left(1-\frac{1}{e}\right)}{\pi^2}} \text{ approx } 0.3578\ \sqrt{L} \tag{16}$$

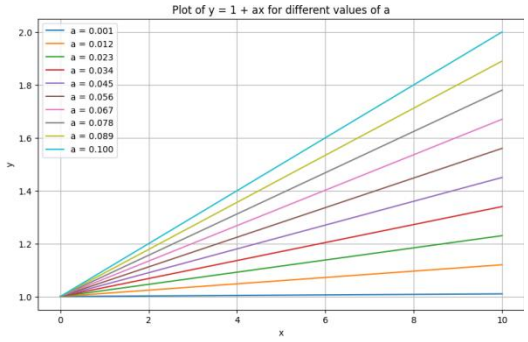

Fig.1 Graph of [y=1+ax] for varying a and x

Discussion

Singular Perturbation Solution.
In the context of the Cahn-Hilliard equation with a double-well potential $f(\phi)=\frac{1}{4}(\phi^2-1)^2$, the equation takes the form:

$$\frac{\partial\phi}{\partial t}=\nabla\cdot\left(M\nabla\left(-\epsilon^2\nabla^2\phi+\frac{\partial f}{\partial\phi}\right)\right) \tag{17}$$

where is the order parameter, M is the mobility, and ε (epsilon) is a small parameter related to the interface width.
A singular perturbation approach involves rescaling the spatial and/or temporal variables to exploit the small parameter epsilon. The idea is to separate the solution into different regions: an inner region where rapid changes occur (like at an interface) and an outer region where the solution varies slowly.

Inner Region. In the inner region, we rescale the spatial variable as $\xi = \frac{x}{\epsilon}$. The leading-order equation in the inner region typically becomes:

$$\frac{d^2\phi}{d\xi^2} = \frac{\partial f}{\partial \phi} \tag{18}$$

with boundary conditions determined by matching the inner solution to the outer solution. For the double-well potential, the solution in the inner region represents the transition layer (interface) between the two phases.

Outer Region. In the outer region, where x is not rescaled, the solution varies slowly, and the terms involving epsilon can often be neglected in the leading order. This results in a simpler equation, typically of the form:

$$\frac{\partial \phi}{\partial t} = M\nabla^2\left(\frac{\partial f}{\partial \phi}\right) \tag{19}$$

Regular Perturbation Solution.A regular perturbation approach assumes that the solution can be expanded $\phi = \phi_0 + \epsilon\phi_1 + \epsilon^2\phi_2 + \cdots.$ (20)

This method works well when the solution is smooth and there are no regions with rapid variations or steep gradients. Each term in the series is obtained by substituting the expansion into the original equation and solving the resulting hierarchy of equations.

Differences and Applications. Singular Perturbation is useful when the solution exhibits layers or interfaces with rapid changes,Regular Perturbation method is appropriate when the solution is smooth and varies slowly, without sharp transitions. It is easier to apply but fails to capture the behavior near interfaces accurately.

Singular perturbation applies when the small parameter ε in (20) significantly affects the behavior of the solution and/or the solution exhibits regions with different scales (e.g., a thin interface versus a bulk phase).
Singular perturbation is essential for accurately solving problems involving phase transitions and interfaces, such as those described by the Cahn-Hilliard equation with a double-well free energy potential. Regular perturbation is simpler but limited to cases where the solution does not exhibit sharp transitions.To apply the singular perturbation method to the Cahn-Hilliard (CH) equation using ε (epsilon) as the small parameter, we start again with the CH equation:

$$\frac{\partial \phi}{\partial t} = \nabla \cdot \left(M\nabla\left(-\epsilon^2\nabla^2\phi + \frac{\partial f}{\partial \phi}\right)\right) \tag{21}$$

where $f(\phi) = \frac{1}{4}(\phi^2 - 1)^2$

Step 1: Inner and Outer Expansions. We expect different behavior in the inner region (near the interface) and the outer region (away from the interface).
Inner Region (Interface)
In the inner region, we introduce the scaled coordinate $\xi = \frac{x}{\epsilon}$. The order parameter ϕ is expanded as:

$$\phi = \phi_0(\xi) + \epsilon\phi_1(\xi) + \epsilon^2\phi_2(\xi) + \cdots \tag{22}$$

Substituting into the CH equation and considering leading-order terms, we get:

$$\frac{d^2\phi_0}{d\xi^2} = \frac{\partial f}{\partial \phi}_{\phi=\phi_0}$$

This simplifies to: $\frac{d^2\phi_0}{d\xi^2} = \phi_0(\phi_0^2 - 1)$ (23)

This is a well-known equation describing the profile of the order parameter across the interface. The solution is:

$$\phi_0(\xi) = \tanh\left(\frac{\xi}{\sqrt{2}}\right)$$

This represents the smooth transition layer between the two phases.

Outer Region (Bulk). In the outer region, where variations are slow, the small parameter \(\epsilon \) does not play a significant role in the leading order. Thus, the equation simplifies to:

$$\frac{\partial \phi}{\partial t} = M\nabla^2\left(\frac{\partial f}{\partial \phi}\right) \quad (24)$$

where we take $(\phi = \phi_0(x) + \epsilon\phi_1(x) + \cdots and\ epsilon \to 0$

Step 2: Matching Conditions. The inner and outer solutions must match in the overlap region. For the leading-order terms, we require that:

$$\lim_{\xi \to \pm\infty} \phi_0(\xi) = \phi_{\text{outer}}(x)$$

This implies that far from the interface, the inner solution matches the outer solution.

Step 3: Composite Solution. The composite solution combines the inner and outer solutions to provide a uniform approximation across the entire domain. It is given by:

$$\phi \approx \phi_{\text{outer}}(x) + \left[\phi_0\left(\frac{x - x_0}{\epsilon}\right) - \phi_{\text{outer}}(x_0)\right] \quad (25)$$

where x_0 is the location of the interface. The expansion of tanh(x/ε√2) is given from Wolfram as

$$\frac{x}{\sqrt{2}\,\epsilon} - \frac{x^3}{6\left(\sqrt{2}\,\epsilon^3\right)} + \frac{x^5}{30\sqrt{2}\,\epsilon^5} - \frac{17\,x^7}{2520\left(\sqrt{2}\,\epsilon^7\right)} + \frac{31\,x^9}{22\,680\sqrt{2}\,\epsilon^9} - \frac{691\,x^{11}}{2\,494\,800\left(\sqrt{2}\,\epsilon^{11}\right)} + O\left(x^{12}\right)$$

(Taylor series) (26)

Results

The singular perturbation approach for the CH equation with epsilon ε as a small parameter reveals that:

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Inner Region (Interface): The order parameterϕ transitions smoothly across the interface according to: $\phi_0(\xi) = \tanh\left(\frac{\xi}{\sqrt{2}}\right)$

Outer Region (Bulk): Away from the interface, the order parameter satisfies the simpler diffusion equation:

$$\frac{\partial \phi}{\partial t} = M\nabla^2\left(\frac{\partial f}{\partial \phi}\right) \quad (27)$$

Composite Solution: Combines inner and outer solutions to provide a uniform approximation.

This approach highlights how singular perturbation methods can effectively capture the behavior of the order parameter near the interface and in the bulk, providing a comprehensive understanding of the solution to the Cahn-Hilliard equation. Figure 2 and 3 illustrate the trend of the solutions.

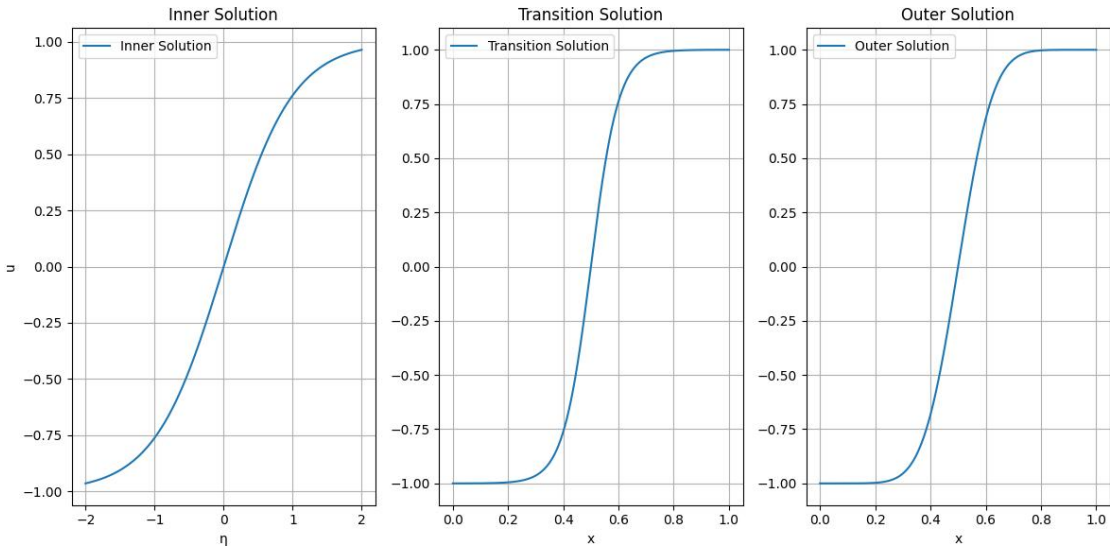

Fig 2 Inner , transition and outer solutions

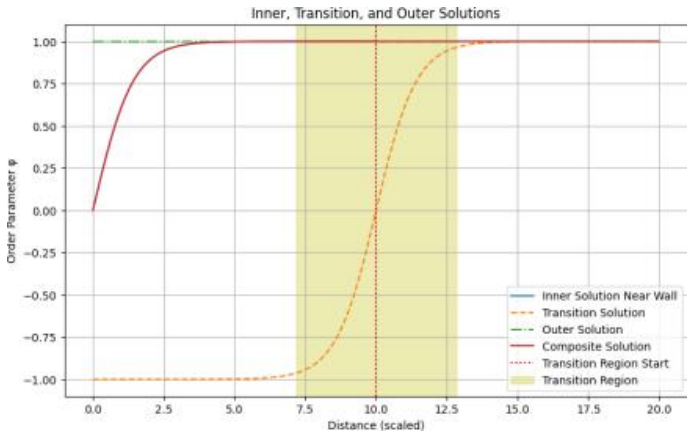

Fig 3. Solutions for large x

Time Dependence in Coarsening. The perturbative solutions for the Cahn-Hilliard equation offer a detailed analysis of phase separation dynamics in thin films. These solutions provide insights into how concentration profiles evolve over time, which is critical for understanding coarsening dynamics. Time Dependence in Coarsening, or Ostwald ripening, is the process where larger domains grow at the expense of smaller ones, leading to a reduction in the total interfacial area and a minimization of the system's free energy. This phenomenon is inherently time-dependent and can be

described by the scaling laws derived from the Cahn-Hilliard equation. Coarsening can be understood through the analysis of perturbative solutions in the following ways:

1. Early Time Regime. At early times, the concentration fluctuations grow exponentially, as captured by the linear stability analysis of the Cahn-Hilliard equation. The perturbative solution shows that for very small times τ, the first few terms dominate, giving a good approximation of the solution.
The initial growth rate of these fluctuations can be expressed

$$u(\tau) \approx (a+b)/2 + (a-b)/2(1 - M(\pi/L)^2\tau)$$

indicating a rapid increase in the concentration differences at small times.

2. Intermediate Time Regime. As time progresses, non-linear effects become significant, and the system transitions into a regime where the domain sizes grow following a power-law behavior. The perturbative approach reveals that higher-order terms decay rapidly, highlighting the transition from linear to non-linear growth.
- This is consistent with the well-known Lifshitz-Slyozov-Wagner (LSW) theory, where the average domain size R grows as $R \sim t^{1/3}$. The paper's findings align with this theory, emphasizing that the higher-order terms' rapid decay leads to a regime where the coarsening follows a predictable scaling law.

3. Late Time Regime. At later times, the coarsening process slows down as the system approaches equilibrium. The perturbative solutions indicate that the concentration profile evolves towards a steady-state solution, where the interface width and domain size stabilize.
- In this regime, the growth of domain size can be influenced by the mobility M and the interfacial width parameter ϵ. The results suggest that the domain growth continues to follow a power-law behavior, albeit with a slower rate due to the diminishing driving force for further coarsening.

Comparison with Precipitation Hardening. The coarsening dynamics discussed above share similarities with the processes observed in precipitation hardening. In both cases, the microstructural evolution is driven by the reduction of free energy, leading to the growth of certain features (domains or precipitates) at the expense of others. The time dependence of these processes can be described using similar scaling laws, highlighting the universal nature of coarsening phenomena in different material systems.

Discussion of Key References. The foundational work by J. W. Cahn and J. E. Hilliard, titled "Free Energy of a Nonuniform System. I. Interfacial Free Energy" (1958), is seminal in the study of phase separation and coarsening dynamics. Their introduction of the Cahn-Hilliard equation provides a theoretical framework to describe how interfacial free energy drives the evolution of concentration profiles in nonuniform systems, laying the groundwork for numerous subsequent studies in materials science and thermodynamics . P. W. Voorhees' paper, "The theory of Ostwald ripening" (1985), complements the Cahn-Hilliard framework by focusing on the kinetics of phase separation, particularly the coarsening process where larger domains grow at the expense of smaller ones. This work is crucial for understanding the time dependence of domain growth during the late stages of phase separation,

providing a theoretical basis that is highly relevant for interpreting perturbative solutions of the Cahn-Hilliard equation .A. Novick-Cohen's review, "The Cahn-Hilliard Equation: Mathematical and Modeling Perspectives" (1998), offers a detailed exploration of the mathematical techniques and modeling applications associated with the Cahn-Hilliard equation. This paper discusses various methods to solve the equation and highlights its broad applicability in simulating phase separation phenomena, making it a valuable resource for researchers applying the Cahn-Hilliard framework to complex systems .K. Glasner's "A boundary integral method for the Cahn-Hilliard equation" (2001) addresses the computational challenges of solving the Cahn-Hilliard equation, particularly in systems with complex geometries. By introducing a numerical approach using boundary integral methods, Glasner's work significantly enhances the ability to simulate phase separation in practical scenarios, contributing to the computational modeling of materials [4].

The study by S. M. Allen and J. W. Cahn, "Ground State Structures in Ordered Binary Alloys with Second Neighbor Interactions" (1972), extends their earlier theoretical work to explore the energetics and stability of ordered phases in binary alloys. This research is critical for predicting microstructural evolution during phase separation, linking the theoretical aspects of the Cahn-Hilliard equation with practical observations in alloy systems [5]

L. Q. Chen's review on "Phase-field models for microstructure evolution" (2002) provides a comprehensive overview of the methodologies used to simulate microstructural evolution in materials. The phase-field approach, which includes the Cahn-Hilliard equation, serves as a powerful tool for studying the dynamics of phase transformations, emphasizing its importance in predicting material behavior and guiding the design of new materials with tailored properties ,[6].

More recent advances in the understanding and application of the Cahn-Hilliard equation are discussed in works such as H. Wu's review (2022), which covers classical results and recent advances in dynamic boundary conditions, [7] , and A. Bonfoh, M. Grasselli, and A. Miranville's "Singularly perturbed 1D Cahn-Hilliard equation revisited" (2010), which revisits and provides new insights into the singular perturbations of the equation .[8] . A. Miranville and S. Zelik's studies (2011, 2009) further explore the Cahn-Hilliard equation with logarithmic potentials and dynamic boundary conditions, expanding its theoretical and practical applications.[9] . Lastly, the comprehensive overview provided by A. Miranville in "The Cahn-Hilliard Equation: Recent Advances and Applications" offers a detailed look into the latest developments and applications of this critical equation in materials science.[10] , while S. O. Londen and H. Petzeltová's work on the regularity and separation from potential barriers for the Cahn-Hilliard equation (2018) adds to the nuanced understanding of the equation's behavior in the presence of singular potentials.[11] .

Conclusion

The perturbative solutions of the Cahn-Hilliard equation provide a comprehensive framework for understanding the time dependence of coarsening in thin films. By examining the evolution of concentration profiles, the paper elucidates the early, intermediate, and late time regimes of coarsening, drawing parallels to the scaling laws observed in precipitation hardening. This analysis not only enhances our

understanding of phase separation dynamics but also offers valuable insights into the broader context of microstructural evolution in materials.

Future Work

Future research could extend this perturbative approach to two and three-dimensional systems, consider the effects of varying mobility M, and incorporate more complex free energy potentials. Numerical simulations could also validate the analytical predictions and explore the nonlinear regime of the Cahn-Hilliard equation.

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