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High accuracy solutions to energy gradient flows from material science models

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ABSTRACT

A computational framework is presented for materials science models that come from energy gradient flows. The models of interest lead to the evolution of structure involving two or more phases. The framework includes higher order derivative models and vector problems. Solutions are considered in periodic cells and standard Fourier spectral discretization in space is used. Implicit time stepping is used with adaptivity based on local error estimates. The implicit system at every time step is solved iteratively with Newton's method. The resulting linear systems are solved in inner iterations with the conjugate gradient method, using a novel preconditioner that is a constant coefficient version of the system, taking values for the coefficients at the pure phase states. Solutions with high spatial and temporal accuracy are obtained. The dependence of the condition number of the preconditioned system on the size of the time step and the order parameter in the model (that represents the scaled width of transition layers between phases) is investigated numerically and with formal asymptotics in a simple setting. The asymptotic results require a conjecture on the rank of a modified square distance matrix. Results from a fast, graphical processing unit implementation for a three-dimensional model are shown. A comparison to time stepping with operator splitting (into convex and concave parts that guarantees energy decrease in the numerical scheme) is done.

1. Introduction

Many material science problems require an understanding of the microstructure that develops in a mixture of two of more materials or phases over time as it phase separates during a casting or annealing process. One such equation is the well-studied Cahn–Hilliard [9] equation, written below in Eq. (2) in a one-dimensional (1D) setting, that describes a binary alloy during annealing. The problem is described by a scalar function u of space and time that takes values $u = +1$ in one phase and $u = -1$ in the other phase. There is a parameter ϵ in the model that describes the width of the layers between the regions. Such regions form in $O(1)$ time in a *spinodal* evolution. Subsequently, they merge in a *ripening* process. Ripening happens on longer time scales, generically $O(e^{C/\epsilon})$ for 1D Cahn–Hilliard [44] and $O(1/\epsilon)$ in higher dimensions [33]. We extend the use of the terms "spinodal" and "ripening" to describe similar regimes in the evolution described by other equations. Phase regions undergoing Cahn–Hilliard evolution increase in size over time in a coarsening process. The statistics of this coarsening process are of interest [7]. The Cahn–Hilliard model is a sub-class of phase field models. A review of

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the use of such models in material science applications can be found in [11]. It can be shown rigorously that as $\epsilon \to 0$, solutions of Cahn–Hilliard equations have layers that tend to interfaces that move with a nonlocal geometric motion known as the Mullins–Sekerka flow [33]. Other phase field models also limit to geometric motion of other kinds. Understanding the limiting process and studying it directly is of interest. In addition, Cahn–Hilliard equations and variants can be used in computational approximation of moving interfaces in so-called *diffuse interface methods* [52,3] in which the problem for u is coupled to other variables describing other physics. While the computational approach developed in this paper might be useful to some diffuse interface computations, we are motivated by a general class of pure (uncoupled to other physics) energy gradient phase field problems described below.

There are several interesting generalizations of the Cahn–Hilliard equation. A lower order version (two instead of four spatial derivatives), the Allen–Cahn equation (1) [2] is also of interest in materials science, describing the evolution of crystal grains of the same material during annealing. This equation can also be called a Ginzberg–Landau equation. Vector versions of this model can describe how these grains can meet at triple junctions [8]. Fourth order phase field models of increasing complexity are used to describe some aspects of cancerous tumour growth [49,15].

Higher derivative equations are also of interest. A sixth order problem is considered in [48] for a regularization of a strongly anisotropic Cahn–Hilliard problem. Another sixth order problem is considered in [31] for the phase field crystal equation. Sixth order models also arise in the study of network formation in functionalized polymers [24]. In these materials, acid side chains are added to hydrophobic polymers. The ionic interaction of these acid groups with water promotes the formation of phase interface, a phenomena different from those observed in the Cahn–Hilliard model. Pore network structures can form in these materials which are of great interest in many applications, including membranes in energy conversion devices such as fuel cells [34]. These materials can be described by the recently proposed sixth order functionalized Cahn–Hilliard equation [37,24,23,16,35]. A simple example is given in Eq. (6) below. Considering [24] in particular, it is clear that there are a wide range of parameters and energy terms that need to be explored to relate these models to particular materials. The extension to vector phase fields is of vital interest to models of mixtures with different wettabilities, or to mixtures undergoing phase change, such as crystallization. The aim of this paper is to develop a numerical approach that can be applied to the wide range of problems above that can easily be adapted to new terms, higher order problems, and extension to vector solutions. It should be made clear we do not attempt to outperform well-developed codes with space and time adaptivity with fast, multi-grid solvers that have been developed for particular problems such as [48,36]. Rather, we develop a reasonably fast time-adaptive technique with general applicability.

Since many questions of interest in materials science are about the microstructure of a bulk material far from boundaries, it is reasonable to consider problems in periodic domains. We use a Fourier spectral discretization which is a natural choice in this setting. Although this does rule out spatial adaptivity, it does admit a fast implementation on Graphical Processing Units (GPU) in the computational framework we develop. We discretize in time using Backward Differentiation Formula (BDF) methods [30] of low order, which have good stability properties. Temporal error estimation is done with Adams–Bashforth (AB) [29] predictors. Newton's method is used to solve the resulting nonlinear problems. The Jacobian matrix in the solve for the Newton update is symmetric since it is the second variational derivative of an energy functional. It is also positive definite for time steps small enough (this is discussed in more detail below). Although the Jacobian is dense for spectral discretizations, multiplication by the matrix can be done quickly using the Fast Fourier Transform (FFT). This motivates our use of the conjugate gradient method [45] to solve the Newton updates. Such an approach used on high order problems requires an efficient preconditioner. We use a constant coefficient version of the problem that is a linearization at pure phase states, which will dominate the solution during ripening. This idea is similar to that used in [14] in fixed point iterations for time stepping for Cahn–Hilliard with operator splitting. Efficient performance is seen with our approach for a wide selection of scalar and vector problems from second to sixth order. Mild increase is seen in preconditioned conjugate gradient (PCG) iteration counts per time step as the time step is increased and ϵ is decreased. Exploration of the performance of the method specifically for the 1D Cahn–Hilliard problem, which has a well understood structure during ripening, shows that the number of PCG iterations per solve scales as $O(\sqrt{k/\epsilon})$ for large time steps k and small ϵ, independent of the spatial discretization.

There have been many other contributions to the numerical solution of the Cahn–Hilliard and related equations. The authors are not aware of a comprehensive review of this topic, but representative examples can be found in [19,22,27,28, 39,46,50,51,48,25,14]. Our work is novel in four ways: we exploit the symmetry of the Jacobian matrix for the fully implicit time stepping problem in a CG method; we propose and analyze the preconditioner for this Jacobian solve and show that it is effective for a number of problems; we implement the method in modern GPU architecture and get fast performance; we demonstrate that the Jacobian matrix is not singular for large time steps during ripening and that fully implicit time stepping leads to accurate solutions with these large time steps with energy decrease.

We discuss further the issue of time stepping. For arbitrary discrete initial data, the fully implicit time discretization problem is known to have a unique solution only when the time step is small enough [17]. As the spatial grid size $h = \Delta x$ is reduced and the initial data on the refined grid is again allowed to be arbitrary, the time step that guarantees unique solutions to the implicit time discretization problem is reduced. In addition, it is not possible in general to show that a fully implicit time step leads to a decrease in the underlying energy. Guarantees of solvability for any time step k and energy decrease are possible for some models with an operator splitting approach due to Eyre [21]. Although never published, this work has been very influential and some of the results are summarized in [42]. Several of the computational approaches cited above use variants of this splitting approach. Some of the splitting techniques lead to nonlinear problems and we show