Efficient linear schemes for the nonlocal Cahn–Hilliard equation of phase field models

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\textbf{A B S T R A C T}

In this paper, we develop two second-order in time, linear and unconditionally energy stable time marching schemes for solving the nonlocal Cahn–Hilliard phase field model. The main challenge to construct efficient and unconditional energy stable schemes for this model is how to design proper temporal discretizations for the nonlocal term that is induced from a convolutional type potential and the nonlinear cubic term that is induced from the double-well bulk potential. We solve these issues by developing two efficient time-stepping schemes using the Invariant Energy Quadratization approach. Its novelty is that all nonlinear terms can be treated semi-explicitly to produce linear schemes. We further show the well-posedness of the resulting linear system as well as its unconditional energy stability rigorously. Various numerical simulations are presented to demonstrate the stability, accuracy, and efficiency of the proposed schemes.

\section{1. Introduction}

In this paper, we aim to develop some efficient and effective numerical schemes for solving the nonlocal Cahn–Hilliard equation. The Cahn–Hilliard equation is an efficient modeling and numerical tool to resolve the motion of free interfaces between multiple material components. See \cite{1–7} and the references therein. The local Cahn–Hilliard model can actually be viewed as the approximation of the nonlocal model where the nonlocal convolutional potential is replaced by differential terms \cite{8,9}. The Cahn–Hilliard equation has been widely used in many fields ranging from physics and materials science to biology, finance and image processing (cf. \cite{7,10–21}).

The intriguing advantage of phase field modeling is that the governing system is usually energy stable and well-posed due to the adopted energy-based variational approach, which provides substantial theoretical and practical support to carry out effective mathematical/numerical analysis and perform reliable computer simulations. For algorithms design, a significant goal is to verify the energy stable property at the discrete level irrespectively of the coarseness of the temporal or spatial discretizations. These kinds of algorithms are called unconditionally energy stable. A scheme with this property is especially preferred for solving stiff systems since the dynamics of coarse-graining (macroscopic) process may undergo rapid changes near the interface, the noncompliance of energy dissipation laws may lead to spurious numerical solutions if the mesh or time step size is not carefully controlled. Thus the unconditional energy stability is not only critical for the numerical scheme to capture the correct long-time dynamics of the system, but also provides sufficient flexibility for dealing with the stiffness issue. We also emphasize that the “unconditional” here means the schemes have no constraints for the time step only from the stability concern. It does not mean that any arbitrarily large time step can be chosen for computations since larger time step size will inevitably induce larger numerical errors in practice. Undoubtedly, higher order time marching schemes are preferable to lower order schemes if the adopted time step is expected to be as large as possible under certain accuracy requests. This fact motivates us to develop higher order schemes, e.g., the second order time marching schemes while preserving the unconditional energy stability in this paper.

It is remarkable that most developments regarding time marching schemes had been focused on the local Cahn–Hilliard equation. Some popular methods include, for instance, the so-called convex splitting approach \cite{22–26}, or the linear stabilized approach \cite{27–34}. About their pros and cons, we give some detailed discussions in Section 3. For the nonlocal Cahn–Hilliard equation, the efforts about algorithm developments or numerical analysis are even more scarce due to the difficulties induced by the convolutional potential. Most of the schemes developed (cf. \cite{35–43}) have used either implicit or explicit time stepping, but it is questionable about
whether they are uniquely solvable or unconditionally energy stable. To the best of the authors’ knowledge, the only unconditionally energy stable scheme was developed in [23,24,44] recently based on the convex splitting approach, where the convex part of the free energy potential is treated implicitly while the concave part is treated explicitly. However, the computational cost is relatively expensive due to its nonlinear nature.

Therefore, the main purpose of this paper is to develop some more efficient and accurate time marching schemes for solving the nonlocal Cahn–Hilliard model. We adopt the Invariant Energy Quadratization (IEQ) approach, which is a novel method and has been successfully applied to solve a variety of gradient flow models (cf. [45–51]) and hydrodynamic models (cf. [52–55]). Its essential idea is to transform the bulk potential into a quadratic form (since the nonlinear potential is usually bounded from below) in a set of new variables via the change of variables. Then, for the reformulated model in terms of the new variables that still retains the identical energy dissipation law, all nonlinear terms can be treated semi-explicitly, which then yields a linear system. As a result, we develop two efficient schemes including the second order Adam–Bashforth and the second order Crank–Nicolson schemes. As shown in details, they are accurate (second order in time), easy-to-implement (linear system), and unconditionally energy stable (with a discrete energy dissipation law). We rigorously prove their unconditional energy stabilities and the well-posedness for the resulted linear systems. Through various numerical simulations, we demonstrate stability and accuracy of the proposed schemes. In addition, although the nonlocal Allen–Cahn equation is not considered in this paper, all proposed numerical schemes can be easily applied to it in a similar way.

The remainder of the paper is organized as follows. In Section 2, we describe the nonlocal Cahn–Hilliard model and its energy dissipative law. In Section 3, we develop a set of numerical schemes and prove their well-posedness and unconditional energy stabilities. In Section 4, we present various numerical simulations to validate the accuracy and stability of the proposed schemes. Finally, some concluding remarks are presented in Section 5.

2. The nonlocal Cahn–Hilliard Equation

We first introduce some notations. The computed domain is \( \Omega = [0, L]^d, d = 2, 3 \). We denote by \( \langle f, g \rangle = \int_{\Omega} f(x)g(x)dx \) the \( L^2 \) inner product between functions \( f(x) \) and \( g(x) \), by \( \| f \| = \sqrt{\langle f, f \rangle} \) the \( L^2 \) norm, by \( \| f \|_{H^1} \) the \( H^1 \) norm, and by \( \| f \|_{L^\infty} \) the \( L^\infty \) norm of a function \( f(x) \), respectively.

2.1. Model formulation

We now give a brief introduction to the nonlocal Cahn–Hilliard model. Supposing \( \phi(x) : \Omega \rightarrow \mathbb{R} \) is a scalar function, the total phenomenological free energy is

\[
E(\phi) = \int_{\Omega} L(\phi(x))dx + \int_{\Omega} F(\phi(x))dx, \tag{1}
\]

where \( L(\phi(x)) \) is a nonlocal convolutional type potential representing the long-range interactions, and \( F(\phi(x)) \) is the local homogeneous energy potential representing the bulk effect. In particular, we focus on the following forms in this paper. \( L(\phi) \) is defined as (cf. [23,24])

\[
L(\phi(x)) = \frac{1}{4} \int_{\Omega} |J(x - y)(\phi(x) - \phi(y))|^2dy, \tag{2}
\]

where \( J : \mathbb{R}^d \rightarrow \mathbb{R} \) is the interaction kernel that is \( \Omega \)-periodic and satisfies the following property:

(i) \( J(-x) = J(x) \), (ii) \( J(x) \geq -B_0 \) for some \( B_0 > 0 \); and the bulk potential \( F(\phi) \) takes the fourth order double well polynomial form as

\[
F(\phi) = \frac{1}{4}(\phi^2 - \gamma_0)^2, \tag{4}
\]

where \( \gamma_0 \) can be an arbitrary real number.

The evolution equation for the phase field variable \( \phi \) is given in a relaxation dynamical form obtained by computing the variational derivatives of the energy in \( H^1 \), as follows:

\[
\phi_t = \nabla \cdot (M(\phi)\nabla \mu), \tag{5}
\]

\[
\mu = f(\phi) - J * \phi + J_0 \phi, \tag{6}
\]

with periodic boundary conditions, where

\[
f(\phi) = F'(\phi) = \phi(\phi^2 - \gamma_0), \quad J * \phi(x) = \int_{\Omega} J(x - y)\phi(y)dy, \tag{7}
\]

\[
J_0 = \int_{\Omega} J(x)dx,
\]

and \( M(\phi) \) is the mobility function. To avoid the degeneracy caused by \( M(\phi) \), we assume that there exist two positive constants \( M_1, M_2 \) such that \( 0 < M_1 \leq M(\phi) \leq M_2 \).

One can easily obtain the energy dissipation property for the model described above. More precisely, by taking the \( L^2 \) inner product of (5) with \( -\mu \) and (6) with \( \phi \), performing integration by parts and summing up the two equalities, we obtain

\[
\frac{d}{dt} E(\phi) = -\| \sqrt{M(\phi)} \nabla \mu \|^2 \leq 0. \tag{8}
\]

We remark that the Cahn–Hilliard type dynamical system (5)–(6) conserves the total volume of the phase variable \( \phi \), i.e., \( \int_{\Omega} \phi dx \). As a matter of fact, by taking the \( L^2 \) inner product of (5) with 1, using integration by parts, one can obtain directly

\[
\frac{d}{dt} \int_{\Omega} \phi(x)dx = 0. \tag{9}
\]

Similarly, the nonlocal Allen–Cahn equation takes the form as

\[
\phi_t = -M(\phi)(f(\phi) - J * \phi + J_0 \phi), \tag{10}
\]

with the corresponding energy law as

\[
\frac{d}{dt} E(\phi) = -\| \sqrt{M(\phi)} \mu \|^2 \leq 0. \tag{11}
\]

To clarify the difference between the local (classical) Cahn–Hilliard model and the nonlocal model (5)–(6), we present the local model here for reference. It reads as

\[
\phi_t = \nabla \cdot (M(\phi)\nabla \mu), \tag{12}
\]

\[
\mu = -\epsilon^2 \Delta \phi + f(\phi), \tag{13}
\]

with the associated energy dissipative law as

\[
\frac{d}{dt} E_{loc}(\phi) = -\| \sqrt{M(\phi)} \mu \|^2 \leq 0, \tag{14}
\]

where

\[
E_{loc}(\phi) = \int_{\Omega} \left( \frac{\epsilon^2}{2} |\nabla \phi|^2 + F(\phi(x)) \right)dx. \tag{15}
\]

We observe that the difference between the local Cahn–Hilliard equation and the nonlocal Cahn–Hilliard equation is in their free energies.

2.2. Intuitive relations between the local free energy and the nonlocal free energy

Here we give some intuitive explanations on the relations between the free energy of the local model (15) and that of the nonlocal model (1).

...
The nonlocal free energy (1) can be written as
\[
E(\phi) = -\frac{1}{2} \int_{\Omega} \int_{\Omega} J(y) \phi(x) \phi(x - y) dxdy + \int_{\Omega} \left( \frac{1}{4} \int_{\Omega} J(y) dy \right) \phi(x)^2 dx + \int_{\Omega} F(\phi(x)) dx,
\]
by noticing the truth
\[
- \frac{1}{2} \int_{\Omega} \int_{\Omega} J(y) \phi(x) \phi(x - y) dxdy = - \frac{1}{4} \int_{\Omega} \int_{\Omega} J(x - y) (\phi(x) - \phi(y))^2 dxdy.
\]
Next, we expand \( \phi(x - y) \) into Taylor series
\[
\phi(x - y) = \sum_{n=0}^{\infty} \frac{(-y \cdot \nabla)^n}{n!} \phi(x).
\]
Substituting (18) into (16), the free energy is casted into
\[
E(\phi) = \int_{\Omega} \left[ - \frac{1}{2} \frac{\phi(x)}{J(y)} \sum_{n=0}^{\infty} \frac{(-y \cdot \nabla)^n \phi(x)}{(2n)!} dy 
+ \frac{1}{2} \left( \int_{\Omega} J(y) dy \right) \phi(x)^2 + F(\phi(x)) \right] dx.
\]
Since \( J(y) = J(-y) \), we have
\[
E(\phi) = \int_{\Omega} \left[ \frac{1}{4} \int_{\Omega} J(y) |y|^2 dy \right] |\nabla \phi(x)|^2 + F(\phi(x)) \right] dx.
\]
If we truncate (18) up to \( n = 1 \), we have
\[
E(\phi) \approx \int_{\Omega} \left[ \frac{1}{4} \int_{\Omega} J(y) |y|^2 dy \right] |\nabla \phi(x)|^2 + F(\phi(x)) \right] dx.
\]
If we set \( F(\phi(x)) = \frac{1}{4} \phi^2 - 1 \) and denote \( \frac{s^2}{2} = \frac{1}{4} \int_{\Omega} J(y) |y|^2 dy \), this could give us the widely used Ginzburg–Landau free energy
\[
E(\phi) \approx \int_{\Omega} \frac{s^2}{2} |\nabla \phi|^2 + \frac{1}{4} (\phi^2 - 1)^2 \right] dx.
\]
Similarly, if we truncate (18) up to \( n = 2 \), it will give us
\[
E(\phi) \approx \int_{\Omega} \left[ \frac{1}{48} \int_{\Omega} J(y) |y|^4 dy \right] |\Delta \phi(x)|^2 
+ \left( \frac{1}{4} \int_{\Omega} J(y) |y|^2 dy \right) |\nabla \phi(x)|^2 + F(\phi(x)) \right] dx.
\]
For certain kernels, we could have \( \frac{1}{48} \int_{\Omega} J(y) |y|^4 dy < 0 \) and \( \frac{1}{4} \int_{\Omega} J(y) |y|^2 dy < 0 \), for instance,
\[
J(y) = \frac{\sigma_1}{\sigma_1^2} \exp\left( -\frac{|y|^2}{\sigma_1^2} \right) - \frac{\sigma_2}{\sigma_2^2} \exp\left( -\frac{|y|^2}{\sigma_2^2} \right),
\]
where \( \sigma_1 = 0.1, \sigma_2 = 0.5, \sigma_1 = 0.08 \) and \( \sigma_2 = 0.2 \). Thus, if we set \( F(\phi(x)) = \frac{1}{4} \phi^4 + \frac{\alpha_1}{\sigma_1^2} \phi^2 \), where \( \alpha, \eta \) are certain constant, this free energy recovers the total free energy of the phase field crystal model \([35,56,57]\)
\[
E(\phi) \approx \int_{\Omega} \left[ a_1 |\Delta \phi|^2 - a_2 |\nabla \phi|^2 + \frac{1}{4} \phi^4 + \frac{1 - \varepsilon}{2} \phi^2 \right] dx, \quad a_1 > 0, \quad a_2 > 0.
\]

3. Numerical schemes

We now construct semi-discrete time marching numerical schemes for solving the nonlocal Cahn–Hilliard equations (5)-(6) and prove their unconditional energy stabilities. It will be clear that the energy stabilities of the semi-discrete schemes are also valid in the fully discrete formulation, for instance by spatially structure-preserving discretizations of finite element method or spectral method.

Comparing to the local Cahn–Hilliard model (12)-(13), besides an issue about how to discretize the nonlinear term \( f(\phi) \) induced by the double well potential, another more difficult challenge lies on how to discretize the nonlocal term \( J \phi \). For \( f(\phi) \), since the traditional fully-implicit or explicit discretization can cause some severe time step constraint relating to the interfacial width \([33,58,59]\), many efforts had been attempted to remove this constraint, in which, the nonlinear convex splitting approach \([22-26, 45,60]\), and the linear stabilized approach \([27,30,33,61-63]\) are two well-known efficient methods. Nonetheless, these two techniques are not optimal choices for the nonlocal Cahn–Hilliard model. For instance, the convex splitting approach brings up nonlinear schemes, thus one need some efficient nonlinear solvers and the implementations are not easy \([23,24]\). The stabilized approach is linear so it is efficient and very easy to implement. But, its stability requests the second order derivative of the nonlinear potential is uniformly bounded. Otherwise, one must modify the nonlinear potential so that the boundedness is true, which in turn leads to an awkward situation that such a modification does not make sense if the PDE solution does not follow the maximum principle. Furthermore, its the second order version for the local Cahn–Hilliard equation, shown in \([33]\), is only conditionally energy stable where the time step is controlled by the interfacial width.

Our goal in this paper is to develop some more efficient time marching schemes. More precisely, we expect that the schemes are linear, unconditionally energy stable, and ready for temporal second order. We adopt the Invariant Energy Quadratization (IEQ) approach developed in \([45,50–52,64]\), which enables us to develop desired numerical schemes without worrying about whether the continuous/discrete maximum principle holds or a convexity/concavity splitting exists.

3.1. An equivalent transformed system

The essential idea of applying the IEQ method for the gradient flow model is to define some new variables to transform the energy potential into a quadratic form. However, note \( f(|x|) \) may not always be positive, thus one cannot directly transform the nonlocal potential \( I(|x|) \) into a quadratic form. To this end, we rewrite the total energy into a summation of several parts as follows:
\[
E(\phi) = \int_{\Omega} \frac{1}{2} \lambda \phi(x)^2 dx 
+ \int_{\Omega} \left( L(\phi(x)) + \frac{B}{2} |\phi(x)|^2 + \frac{B}{2} \int_{\Omega} \phi(x)^2 dy dx \right)
+ \int_{\Omega} \left( F(\phi(x)) - B |\Omega| + \frac{1}{2} \lambda \phi(x)^2 \right) dx.
\]
where we add a zero term
\[
\frac{\lambda}{2} \int_{\Omega} \phi(x)^2 dx + \frac{B}{2} |\Omega| \int_{\Omega} \phi(x)^2 dx + \frac{B}{2} \int_{\Omega} \phi(x)^2 dy dx 
- \int_{\Omega} (B |\Omega| + \frac{1}{2} \lambda) \phi(x)^2 dx.
\]
where $\lambda$ and $B$ work as stabilizing parameters, that is a well-known trick used in the linear stabilized approach mentioned above. Notice $J(x)$ satisfies the property of bounded from below, thus we can establish the bounded from below property for part I and part II. We recall that, in some literatures (cf. [65]), the kernel $J(x)$ is assumed to be always positive, for this case we can simply choose $B = 0$ and all theoretical works are still valid.

**Lemma 3.1.** Part I and part II are both bounded from below. More precisely, for part I, we have

$$L(\phi(x)) + \frac{B}{2} |\Omega| |\phi(x)|^2 + \frac{B}{2} \int_{\Omega} \phi(y)^2 \, dy \geq 0,$$

for any $\phi$ and $x \in \Omega$ and $B > B_0$.

**Proof.** For part I, by using $J(x) \geq -B_0 \geq -B$ and $-(a - b)^2 \geq -2(a^2 + b^2)$, we obtain

$$L(\phi(x)) + \frac{B}{2} |\Omega| |\phi(x)|^2 + \frac{B}{2} \int_{\Omega} \phi(y)^2 \, dy \geq - \frac{B}{2} \int_{\Omega} (|\phi(x)|^2 + \phi(y)^2) \, dy + \frac{B}{2} \int_{\Omega} \phi(y)^2 \, dy \geq 0.$$  

For part II, we can find its lower bound as

$$F(x) - (B_0 |\Omega| + \frac{1}{2} \lambda) y^2 \geq \frac{1}{4} |\Omega| (y_0 + 2B_0 |\Omega| + \lambda) y^2. \quad \Box $$

We now introduce two time-dependent scalar auxiliary variables $U$ and $V$ in terms of $\phi$ as follows:

$$U(\phi) = \sqrt{L(\phi(x)) + \frac{B}{2} |\Omega| |\phi(x)|^2 + \frac{B}{2} \int_{\Omega} \phi(y)^2 \, dy + A_1},$$

$$V(\phi) = \sqrt{F(\phi(x)) - (B |\Omega| + \frac{1}{2} \lambda) |\phi(x)|^2 + A_2},$$

where $A_1$ and $A_2$ are two positive constants to ensure the radicands to be positive.

In terms of these two scalar auxiliary variables, the total energy (26) becomes

$$E(\phi, U, V) = \int_{\Omega} \left( \frac{1}{2} |\phi|^2 + |U|^2 + |V|^2 \right) \, dx - A_1 |\Omega| - A_2 |\Omega|.$$  

Then we obtain an equivalent PDE system by taking the time derivative for the new variable $U$, $V$, as follows:

$$\phi_t = \nabla \cdot (M(\phi) \nabla \mu),$$

$$\mu = \lambda \phi + HU + SV,$$

$$U_t = \frac{1}{2} H \phi_t,$$

$$V_t = \frac{1}{2} S \phi_t,$$

where

$$H(\phi) = \frac{-J \ast \phi + J_0 \phi + B |\Omega| \phi + B |\Omega| \int_{\Omega} \phi(x) \, dx}{\sqrt{L(\phi(x)) + \frac{1}{2} |\Omega| |\phi(x)|^2 + \frac{1}{2} \int_{\Omega} \phi(y)^2 \, dy + A_1}},$$

$$S(\phi) = \frac{f(\phi) - 2B |\Omega| \phi - \lambda \phi}{\sqrt{F(\phi(x)) - (B |\Omega| + \frac{1}{2} \lambda) |\phi(x)|^2 + A_2}}.$$  

The boundary conditions are periodic for all variables, and the initial conditions read as

$$\phi|_{t=0} = \phi^0, \quad U|_{t=0} = U(\phi^0), \quad V|_{t=0} = V(\phi^0).$$

The new transformed PDE system (32)–(35) admits an energy dissipative law. In details, by taking the $L^2$ inner product of (32) with $\mu$, of (33) with $-\phi$, of (34) with $2U$ and of (35) with $2V$, then performing integration by parts and summing up all equalities, we obtain

$$\frac{d}{dt} E(\phi, U, V) = - \|M(\phi) \nabla \mu\|^2 \leq 0.$$  

**Remark 3.1.** The new transformed system (32)–(35) is equivalent to the original system (5)–(6) since (30) can be easily obtained by integrating (34) and (35) with respect to the time. Therefore, the new energy law (38) for the transformed system is exactly the same as the energy law (8) for the original system for the time continuous case.

**Remark 3.2.** In numerical practice, we simply choose a relative big constant $A_1$ and set $B = 0$. In addition, the scheme could be simplified given specific expression of $F(\phi)$. For instance, in the numerical simulations, if we consider $F(\phi) = \frac{1}{2} (\phi^2 - \gamma_0)^2$, we can set

$$V(\phi) = \frac{1}{2} (\phi^2 - (2B |\Omega| + \lambda + \gamma_0)), \quad S(\phi) = 2 \phi,$$

by choosing $A_2 = \frac{1}{4} (2B |\Omega| + \gamma_0)^2 - \frac{1}{4} \gamma_0^2$.

The time marching numerical schemes are developed to solve the new transformed system (32)–(35). The proof of the unconditional stability of the schemes follows the similar strategy as in the derivation of the energy law (38).

### 3.2. Adam–Bashforth scheme

We define two Sobolev spaces:

$L^2_{\mu}(\Omega) = \{ \psi \text{ is periodic, } \psi \in L^2(\Omega) \}$ and $\int_{\Omega} \psi = 0$,

$H^1_{\mu}(\Omega) = \{ \psi \text{ is periodic, } \psi \in H^1(\Omega) \}$ and $\int_{\Omega} \psi = 0$.

Let $\delta t > 0$ be a time step size and set $t^n = n \delta t$ for $0 < n < N = [T / \delta t]$, let $\psi^n$ denotes the numerical approximation to $\psi(\cdot, t_{n-1})$, and $\psi^{n+1} = 2 \psi^{n+1} - \psi^n$, for any function $\psi$. We construct a second order Adam–Bashforth type scheme for solving (32)–(35).

**Scheme 1 (Adam–Bashforth Scheme).** Assuming that $\phi^{n-1}, U^{n-1}, V^{n-1}$ and $\phi^n, U^n, V^n$ are known, we update $\phi^{n+1}, U^{n+1}$ and $V^{n+1}$ as follows:

$$\begin{aligned}
3 \phi^{n+1} - 4 \phi^n + \phi^{n-1} &= \nabla \cdot (M(\phi^{n+1}) \nabla \mu^{n+1}), \\
\mu^{n+1} &= \lambda \phi^{n+1} + H \phi^{n+1} U^{n+1} + S \phi^{n+1} V^{n+1}.
\end{aligned}$$

$$
\begin{aligned}
U^{n+1} &= H \phi^{n+1} U^{n+1} + S \phi^{n+1} U^{n+1} + g^n, \\
V^{n+1} &= S \phi^{n+1} V^{n+1} + g^n.
\end{aligned}$$

where

$$
\begin{aligned}
3 \phi^{n+1} - 4 \phi^n + \phi^{n-1} &= \nabla \cdot (M(\phi^{n+1}) \nabla \mu^{n+1}), \\
\mu^{n+1} &= \lambda \phi^{n+1} + H \phi^{n+1} U^{n+1} + S \phi^{n+1} V^{n+1}.
\end{aligned}$$

The boundary conditions for all variables are periodic.

Note that the nonlinear coefficient $H \phi^{n+1}$ and $S \phi^{n+1}$ of the new variables $U$ and $V$ are both treated explicitly, therefore we can rewrite (42) and (43) as

$$
\begin{aligned}
U^{n+1} &= H \phi^{n+1} U^{n+1} + g^n, \\
V^{n+1} &= S \phi^{n+1} V^{n+1} + g^n.
\end{aligned}$$

where

$$
\begin{aligned}
g^n_1 &= \frac{4U^n - U^{n-1}}{3} - \frac{1}{2} H \phi^{n+1} 4 \phi^n - \phi^{n-1}, \\
g^n_2 &= \frac{4V^n - V^{n-1}}{3} - \frac{1}{2} S \phi^{n+1} 4 \phi^n - \phi^{n-1}.
\end{aligned}$$

To rewrite (42) and (43) as

$$
\begin{aligned}
U^{n+1} &= H \phi^{n+1} U^{n+1} + g^n, \\
V^{n+1} &= S \phi^{n+1} V^{n+1} + g^n.
\end{aligned}$$

where

$$
\begin{aligned}
g^n_1 &= \frac{4U^n - U^{n-1}}{3} - \frac{1}{2} H \phi^{n+1} 4 \phi^n - \phi^{n-1}, \\
g^n_2 &= \frac{4V^n - V^{n-1}}{3} - \frac{1}{2} S \phi^{n+1} 4 \phi^n - \phi^{n-1}.
\end{aligned}$$


By plugging (44) into (41), we obtain an equivalent scheme as follows.

**Scheme 2.** Assuming that \( \phi^{n-1}, U^{n-1}, V^{n-1} \) and \( \phi^n, U^n, V^n \) are known, we update \( \phi^{n+1}, U^{n+1}, V^{n+1} \) from the following two steps.

- **Step 1:** we update \( \phi^{n+1} \) by solving
  \[
  \frac{3\phi^{n+1} - 4\phi^n + \phi^{n-1}}{2\delta t} = \nabla \cdot (M(\phi^{n+1})\nabla \mu^{n+1}),
  \]
  \[
  \mu^{n+1} = \lambda \phi^{n+1} + \frac{1}{2} (H^{n+1} H^{n+1} + S^{n+1} S^{n+1}) \phi^{n+1} + H^{n+1} b^n_1 + S^{n+1} b^n_2.
  \]

  using the periodic boundary conditions.

- **Step 2:** we update \( U^{n+1} \) and \( V^{n+1} \) via (44).

It is remarkable that, in Scheme 2, once \( \phi^{n+1} \) is obtained from (46)–(47), the new variables \( U^{n+1} \) and \( V^{n+1} \) can be automatically updated in (44). Namely, the new variables \( U \) and \( V \) do not involve any extra computational costs, indeed.

We now show the well-posedness of Scheme 2. By taking the \( L^2 \) inner product of (40) with 1, using integration by parts, we derive

\[
\int_{\Omega} \phi^{n+1} dx = \int_{\Omega} \phi^n dx = \cdots = \int_{\Omega} \phi^0 dx.
\]

Let \( \alpha_0 = \int_{\Omega} \phi^0 dx, \beta_{\mu}^{n+1} = \int_{\Omega} \mu^{n+1} dx \), and we define

\[
\phi = \phi^{n+1} - \alpha_0, \mu = \mu^{n+1} - \beta_{\mu}^{n+1}.
\]

Thus, the weak form for (46)–(47) can be written as the following system with the unknowns \( (\mu, \phi) \in (H_{\text{per}}, L^2_{\text{per}})(\Omega) \),

\[
(\phi, w) + \frac{2}{3} \delta t (M(\phi^{n+1})\nabla \mu, \nabla w) = \frac{1}{4} (4\phi^n - \phi^{n-1}, w), w \in H_{\text{per}}.
\]

\[
(-\mu, \psi) + \lambda (\mu, \phi) + \frac{1}{2} (H^{n+1} \psi, H^{n+1} \psi) + \frac{1}{2} (S^{n+1} \phi, S^{n+1} \phi) = (\beta^n_1, \psi), \psi \in L^2_{\text{per}}(\Omega),
\]

where \( \beta^n_1 = -(H^{n+1} b^n_1 + S^{n+1} b^n_2) + \frac{1}{2} \alpha_0 (H^{n+1} b^n_1 + S^{n+1} b^n_2) \).

We denote the above linear system (50)–(51) as

\[
(AX, Y) = (B, Y),
\]

where \( A \) is the linear operator, \( X = (\mu, \phi)^T, Y = (w, \psi)^T, B = \left( \frac{1}{2} (4\phi^n - \phi^{n-1}, b^n_1), \beta^n_1 \right)^T \), and \( X, Y \in (H_{\text{per}}, L^2_{\text{per}})(\Omega) \).

The well-posedness of the above linear system (52) is shown as follows.

**Theorem 3.1.** The linear system (52) admits a unique solution \( (\mu, \phi) \in (H_{\text{per}}, L^2_{\text{per}})(\Omega) \).

**Proof.** (i) For any \( X = (\mu, \phi)^T, Y = (w, \psi)^T \) with \( X, Y \in (H_{\text{per}}, L^2_{\text{per}})(\Omega) \), we have

\[
(AX, Y) \leq C_1 (\|\phi\| + \|\mu\|_1 \|\psi\| + \|w\|_H^1),
\]

where \( C_1 \) depends on \( \delta t, \lambda, M_2, \|H^{n+1}\|_{L^\infty} \) and \( \|S^{n+1}\|_{L^\infty} \). Therefore, the bilinear form \((AX, Y)\) is bounded.

(ii) It is easy to derive that, for any \( X = (\mu, \phi)^T \in (H_{\text{per}}, L^2_{\text{per}})(\Omega) \), we have

\[
(AX, X) = \frac{2}{3} \delta t (M(\phi^{n+1})\nabla \mu, \nabla \mu) + \lambda \|\phi\|^2
+ \frac{1}{2} (H^{n+1} \phi, H^{n+1} \phi) + \frac{1}{2} (S^{n+1} \phi, S^{n+1} \phi)
\geq \frac{2}{3} \delta t \|\nabla \mu\|^2 + \lambda \|\phi\|^2 \geq C_2 (\|\mu\|^2 + \|\phi\|^2),
\]

where \( C_2 \) depends on \( \delta t, \lambda, M_1 \). Thus the bilinear form \((AX, Y)\) is coercive.

Then from the Lax–Milgram theorem, we conclude the linear system (52) admits a unique solution \( (\mu, \phi) \in (H_{\text{per}}, L^2_{\text{per}})(\Omega) \). \( \square \)

The energy stability of Scheme 1 (or Scheme 2) is presented as follows.

**Theorem 3.2.** The scheme (40)–(43) is unconditionally energy stable, and satisfies the following discrete energy dissipation law:

\[
\frac{1}{\delta t} (E_{\text{bef}}^{n+1} - E_{\text{bef}}^{n+1}) \leq -\|\sqrt{M(\phi^{n+1})} \nabla \mu^{n+1}\|^2,
\]

where

\[
E_{\text{bef}}^{n+1} = \frac{\lambda}{4} (\|\phi^{n+1}\|^2 + 2\|\phi^{n+1} - \phi^n\|^2)
+ \frac{1}{2} (\|U^{n+1}\|^2 + 2U^{n+1} - U^n)^2)
+ \frac{1}{2} (\|V^{n+1}\|^2 + 2V^{n+1} - V^n)^2).
\]

**Proof.** By taking the \( L^2 \) inner product of (40) with \(-2\delta t \mu^{n+1} \) and using integration by parts, we obtain

\[
-(3\phi^{n+1} - 4\phi^n + \phi^{n-1}, \mu^{n+1}) = 2\delta t \|\sqrt{M(\phi^{n+1})} \nabla \mu^{n+1}\|^2.
\]

By taking the \( L^2 \) inner product of (41) with \(3\phi^{n+1} - 4\phi^n + \phi^{n-1} \), and applying the following identity

\[
(3a - 4b + c, 2a) = a^2 - b^2 + (2a - b)^2 - (2b - c)^2 + (a - 2b + c)^2.
\]

we can derive

\[
\frac{\lambda}{2} \left( \|\phi^{n+1}\|^2 + 2\|\phi^{n+1} - \phi^n\|^2 - \|\phi^n\|^2 - 2\|\phi^{n+1} - \phi^n\|^2 \right)
\]

\[
+ \frac{\lambda}{2} \|\phi^{n+1} - 2\phi^n + \phi^{n-1}\|^2
\]

\[
+ (H^{n+1} U^{n+1} + S^{n+1} V^{n+1}, 3\phi^{n+1} - 4\phi^n + \phi^{n-1}).
\]

By taking the \( L^2 \) inner product of (42) with \(-2U^{n+1} \) and applying (58), we obtain

\[
-(\|U^{n+1}\|^2 + \|2U^{n+1} - U^n\|^2 - \|U^n\|^2 - 2|U^n - U^{n-1}|^2)
+ \|U^{n+1} - 2U^n + U^{n-1}\|^2
\]

\[
= -(H^{n+1} (3\phi^{n+1} - 4\phi^n + \phi^{n-1}), U^{n+1} + V^{n+1}).
\]

By taking the \( L^2 \) inner product of (43) with \(-2V^{n+1} \) and applying (58), we obtain

\[
-(\|V^{n+1}\|^2 - \|V^n\|^2 + 2|V^{n+1} - V^n|^2)
= -(S^{n+1} (3\phi^{n+1} - 4\phi^n + \phi^{n-1}), V^{n+1}).
\]
Remark 3.3. The discrete energy inequality (55) can ensure the $L^2$ bound for the numerical solution when $\lambda \neq 0$. Actually, the proofs of boundedness and coercivity in Theorem 3.1 depend on Remark 3.3.

Remark 3.4. Heuristically, the $\frac{1}{\delta t}E^{n+1}_\text{(inf)} - E^n_{\text{ref}}$ is a second order approximation of $\frac{d}{dt}E(\phi, u)$ at $t = f^{n+1}$. For instance, for any smooth variable $U$ with time, one can write
\[ \frac{\delta U^{n+1} - U^{n}}{\delta t} = \|U^{n+1} - U^n\| - \|U^n - U^{n-1}\|^2 \]
\[ = \frac{2\delta t}{\delta t} \left( \|U^{n+1}\|^2 + 2\|U^{n+1} - U^n\|^2 - \|2U^n - U^{n-1}\|^2 \right) \]
\[ \approx \left( \frac{\|U^{n+1}\|^2 + \|U^n - U^{n-1}\|^2}{\delta t} \right) + O(\delta t^2) \]
\[ = \frac{d}{dt}(\|U(t^n)\|^2) + O(\delta t^2). \]

3.3. Crank–Nicolson scheme

Alternatively, one can easily develop another second order version based on the Crank–Nicolson type approach as follows.

Scheme 3 (Crank-Nicolson Scheme). Assuming that $\phi^{n-1}$, $U^{n-1}$, $V^{n-1}$ and $\phi^n$, $U^n$, $V^n$ are known, we update $\phi^{n+1}$, $U^{n+1}$, $V^{n+1}$ as follows:

\[ \frac{\phi^{n+1} - \phi^n}{\delta t} = \nabla \cdot (M(\phi^{n+\frac{1}{2}})\nabla \mu^{n+\frac{1}{2}}), \]
\[ \mu^{n+\frac{1}{2}} = \frac{1}{2}(\phi^{n+1} + \phi^n) + H^{n+\frac{1}{2}} \frac{U^{n+1} + U^n}{2} + S^{n+\frac{1}{2}} \frac{V^{n+1} + V^n}{2}, \]
\[ U^{n+1} - U^n = \frac{1}{2}(H^{n+\frac{1}{2}}(\phi^{n+1} - \phi^n)), \]
\[ V^{n+1} - V^n = \frac{1}{2}(H^{n+\frac{1}{2}}(\phi^{n+1} - \phi^n) + H^{n+\frac{1}{2}}(\phi^{n+1} - \phi^n)), \]

where $\phi^{n+\frac{1}{2}} = \frac{1}{2}(\phi^{n+1} - \phi^{n-1})$, $H^{n+\frac{1}{2}} = H(\phi^{n+\frac{1}{2}})$ and $S^{n+\frac{1}{2}} = S(\phi^{n+\frac{1}{2}})$.

By applying the similar process as the Adam–Bashforth scheme, the Crank–Nicolson scheme (64)–(67) is equivalent to the following scheme.

Scheme 4. Assuming that $\phi^{n-1}$, $\phi^n$, $U^n$ and $V^n$ are known, we solve $\phi^{n+1}$, $U^{n+1}$ from the following two steps:

- Step 1: we update $\phi^{n+1}$ by

\[ \frac{\phi^{n+1} - \phi^n}{\delta t} = \nabla \cdot (M(\phi^{n+\frac{1}{2}})\nabla \mu^{n+\frac{1}{2}}), \]
\[ \mu^{n+\frac{1}{2}} = \frac{1}{2}(\phi^{n+1} + \phi^n) + \left(\frac{1}{2}(H^{n+\frac{1}{2}} + S^{n+\frac{1}{2}})\right)\phi^{n+1} + \frac{1}{4}(H^{n+\frac{1}{2}} + S^{n+\frac{1}{2}})\phi^{n+1} + g^n, \]

where $g^n = \frac{1}{2}\phi^n + H^{n+\frac{1}{2}}(U^n - \frac{1}{2}H^{n+\frac{1}{2}}\phi^n) + S^{n+\frac{1}{2}}(V^n - \frac{1}{2}S^{n+\frac{1}{2}}\phi^n)$.

- Step 2: we update $U^{n+1}$ and $V^{n+1}$ by

\[ \begin{align*}
U^{n+1} &= \frac{1}{2}H^{n+\frac{1}{2}}\phi^{n+1} + U^n - \frac{1}{2}H^{n+\frac{1}{2}}\phi^n, \\
V^{n+1} &= \frac{1}{2}S^{n+\frac{1}{2}}\phi^{n+1} + V^n - \frac{1}{2}S^{n+\frac{1}{2}}\phi^n.
\end{align*} \]

The well-posedness of the linear system in the weak form of (68)–(69) can be proved using similar arguments for the Adam–Bashforth scheme, thus we omit the proof here and only show the energy stability as follows.

Theorem 3.3. The scheme (64)–(67) is unconditionally energy stable and satisfies the following discrete energy dissipation law:

\[ \frac{1}{\delta t}(E^{n+1}_\text{en} - E^n_{\text{en}}) = -\frac{1}{2}\nabla \cdot (M(\phi^{n+\frac{1}{2}})\nabla \mu^{n+\frac{1}{2}})^2 \leq 0, \]

where

\[ E^{n+1}_\text{en} = \frac{\lambda}{2}\|\phi^{n+1}\|^2 + \|U^{n+1}\|^2 + \|V^{n+1}\|^2. \]

Proof. By taking the $L^2$ inner product of (64) with $-\delta t \mu^{n+\frac{1}{2}}$, we obtain

\[ -\langle \phi^{n+1} - \phi^n, \mu^{n+\frac{1}{2}} \rangle = \delta t \langle \nabla M(\phi^{n+\frac{1}{2}})\nabla \mu^{n+\frac{1}{2}}, \mu^{n+\frac{1}{2}} \rangle. \]

By taking the $L^2$ inner product of (65) with $\phi^{n+1} - \phi^n$, we can derive

\[ \langle \phi^{n+1} - \phi^n, \mu^{n+\frac{1}{2}} \rangle = \frac{\lambda}{2}(\|\phi^{n+1}\|^2 - \|\phi^n\|^2) + (H^{n+\frac{1}{2}}(\phi^{n+1} - \phi^n), \frac{U^{n+1} + U^n}{2}) \]
\[ + (S^{n+\frac{1}{2}}(\phi^{n+1} - \phi^n), \frac{V^{n+1} + V^n}{2}). \]

By taking the $L^2$ inner product of (66) with $-\frac{U^{n+1} - U^n}{2}$, we obtain

\[ -\langle \|U^{n+1}\|^2 - \|U^n\|^2 \rangle = -\left(H^{n+\frac{1}{2}}(\phi^{n+1} - \phi^n), \frac{U^{n+1} + U^n}{2} \right). \]

By taking the $L^2$ inner product of (67) with $-\frac{V^{n+1} + V^n}{2}$, we obtain

\[ -\langle \|V^{n+1}\|^2 - \|V^n\|^2 \rangle = -\left(S^{n+\frac{1}{2}}(\phi^{n+1} - \phi^n), \frac{V^{n+1} + V^n}{2} \right). \]

By combining (73), (74), (75) and (76), we obtain

\[ \begin{align*}
\frac{\lambda}{2}(\|\nabla \phi^{n+1}\|^2 - \|\nabla \phi^n\|^2) &+ \frac{1}{2}\|U^{n+1}\|^2 - \|U^n\|^2 + \frac{1}{2}\|V^{n+1}\|^2 - \|V^n\|^2 \leq -\delta t \langle \nabla M(\phi^{n+\frac{1}{2}})\nabla \mu^{n+\frac{1}{2}}, \mu^{n+\frac{1}{2}} \rangle.
\end{align*} \]

Finally, we obtain the desired result (71). □

3.4. Spatial discretization

We use the second-order compact finite difference method to discretize the space. Let $N_x, N_t$ be positive integers and the domain $\Omega = [0, L_x] \times [0, L_y]$. We divide the domain into rectangular meshes with mesh size $h_x = L_x/N_x$, $h_y = L_y/N_y$. Following the notations in [54,66,67], we define the following uniform grid:

\[ E := \{x_i \mid i \in \mathbb{Z}\}, \quad C := \{x_i \mid i \in \mathbb{Z}\}, \quad x_i = (i - 1/2)h. \]
Then we can define 2D discrete periodic function spaces:

\[
\mathcal{C} : \{ v : C \times C \rightarrow \mathbb{R} \mid v_{ij} = v_{i+\alpha N_x, j+\beta N_y}, \\
\text{Vi, j, } \alpha, \beta \in \mathbb{Z} \},
\]

\[
\mathcal{E}^{ew} : \{ v : E \times C \rightarrow \mathbb{R} \mid v_{i+1/2,j} = v_{i+\alpha N_x, j+\beta N_y}, \\
\text{Vi, j, } \alpha, \beta \in \mathbb{Z} \},
\]

(79)

\[
\mathcal{E}^{ms} := \{ v : C \times E \rightarrow \mathbb{R} \mid v_{ij+1/2} = v_{i+\alpha N_x, j+1/2 \beta N_y}, \\
\text{Vi, j, } \alpha, \beta \in \mathbb{Z} \}.
\]

The functions \( C \) are called cell-centered functions, and \( \mathcal{E}^{ew}, \mathcal{E}^{ms} \)
are called east–west edge-centered, north–south edge-centered functions, respectively.

We define the center-to-east–west-edge average and difference operator \( A_x, D_x : C \rightarrow \mathcal{E}^{ew} \), and center-to-north–south-edge average and difference operator \( A_y, D_y : C \rightarrow \mathcal{E}^{ms} \) as

\[
D_x \phi_{i,j+1/2} = \frac{1}{h_x} (\phi_{i+1,j} - \phi_{i,j}), \quad A_x \phi_{i+1/2,j} = \frac{1}{2} (\phi_{i+1,j} + \phi_{i,j}),
\]

\[
D_y \phi_{i+1/2,j} = \frac{1}{h_y} (\phi_{i+1,j} - \phi_{i,j}), \quad A_y \phi_{i,j+1/2} = \frac{1}{2} (\phi_{i+1,j} + \phi_{i,j}).
\]

and we define the east–west–center difference and north–south–edge-to-center difference operator \( d_x : \mathcal{E}^{ew} \rightarrow C, d_y : \mathcal{E}^{ms} \rightarrow C \) as

\[
d_x \phi_{i,j} = \frac{1}{h_x} (\phi_{i+1/2,j} - \phi_{i-1/2,j}), \quad d_y \phi_{i,j} = \frac{1}{h_y} (\phi_{i,j+1/2} - \phi_{i,j-1/2}).
\]

Then the spatial discretizations for the Adam–Bashforth scheme read as

\[
\frac{3\phi^{n+1} - 4\phi^n + \phi^{n-1}}{2\delta t} = d_x(A_x(M(\phi^{n+1}))D_x\mu^{n+1}) + d_y(A_y(M(\phi^{n+1}))D_y\mu^{n+1})\big|_{ij},
\]

\[
i = 1, \ldots, N_x, j = 1, \ldots, N_y,
\]

\[
\mu^{n+1} = \lambda \phi^{n+1} + H^{n+1} \phi^{n+1} + S^{n+1} \phi^{n+1} \big|_{ij},
\]

\[
i = 1, \ldots, N_x, j = 1, \ldots, N_y,
\]

(82)

\[
3U^{n+1} - 4U^n + U^{n-1} = \frac{1}{2} H^{n+1} (3\phi^{n+1} - 4\phi^n + \phi^{n-1})\big|_{ij},
\]

\[
i = 1, \ldots, N_x, j = 1, \ldots, N_y,
\]

\[
3V^{n+1} - 4V^n + V^{n-1} = \frac{1}{2} S^{n+1} (3\phi^{n+1} - 4\phi^n + \phi^{n-1})\big|_{ij},
\]

\[
i = 1, \ldots, N_x, j = 1, \ldots, N_y.
\]

The Crank–Nicolson scheme takes the similar form thus we omitted the details here. Moreover, one can readily show that the discrete energy laws of the fully discrete schemes still hold in the same way as in the semi discrete schemes.

**Remark 3.5.** Even for the local Cahn–Hilliard or Allen–Cahn model, to derive the error analysis for the EQ method for the general nonlinear potential is non-trivial. The essential difficulty arises from the way of quadratization for the nonlinear potential \( F(\phi) \) to introduce the new variable, that actually leads the new variable to act as a “encapsulation”, making it difficult to estimate the quantitative relation between the new and original variables. For the nonlinear model, things become more complicated since the nonlinear part is also made to be quadratic by another new auxiliary variable. The related error analysis work, including the semi-discrete scheme and fully-discrete schemes in the context of finite element method or spectral method, is expected to be implemented in the future by following the same line of procedure as [23,24,33,44,68–72].

### 4. Numerical simulations

We now present various numerical simulations to validate the theoretical results derived in the previous section numerically and demonstrate the efficiency, accuracy and stability of the developed schemes. We denote the Adam–Bashforth scheme (40)–(43) by BDF2, and the Crank–Nicolson scheme (64)–(67) by CN2. In all examples, we use the rectangular domain \([0, L_x] \times [0, L_y]\). If not explicitly specified, the default values of parameters are given as follows:

\[
M(\phi) = 1, \quad \gamma_0 = 0.1, \quad A_1 = 1, \quad A_2 = 1,
\]

\[
\lambda = 1, \quad B = 0, \quad L_x = L_y = 5,
\]

which are consistent with the numerical examples performed in the literatures [23,24]. We define the following two nonlocal kernels:

\[
J(x, y, t = 0) = \frac{\alpha}{\sigma_1^2} \exp \left(-\frac{x^2 + y^2}{\sigma_1^2}\right),
\]

(84)

\[
J(x, y) = \frac{\alpha_1}{\sigma_1^2} \exp \left(-\frac{x^2 + y^2}{\sigma_1^2}\right) - \frac{\alpha_2}{\sigma_2^2} \exp \left(-\frac{x^2 + y^2}{\sigma_2^2}\right).
\]

(85)

#### 4.1. Accuracy test

We first test the time convergence rates of the two developed schemes by performing refinement tests. We set the initial conditions as

\[
\phi(x, y, t = 0) = 0.1 \sin \left(\frac{2\pi x}{L_x}\right) \sin \left(\frac{2\pi y}{L_y}\right),
\]

(86)

Use the nonlocal kernel (84) with \( \alpha = 0.1 \) and \( \sigma_1 = 0.08 \) with its profile projection in 1D (by assuming \( x = y \)) in Fig. 1(a).

We perform the refinement test of the time step size. We use \( N_x = N_y = 128 \) grid points. By taking a linear refinement path of \( \delta t = \frac{0.01}{2^k} \) for \( k = 0, 1, \ldots, 6 \), we calculate the rate at which the Cauchy difference, e.g. \( \| \xi^{k} - \xi^{k-1} \|_{L^2} \) varies at the time \( t = 0.2 \) for schemes BDF2 and CN2 in Tables 1 and 2, respectively. We observe these two schemes achieve approximately second order accuracy in time.

#### 4.2. Coarsening dynamics

Next, we study phase separation behavior with the nonlocal Cahn–Hilliard equation. The process of the phase separation can be studied by considering an isotropic binary mixture, which is quenched into the unstable part of its miscibility gap. In this case, the spinodal decomposition takes place, which manifests in the spontaneous growth of the concentration fluctuations that leads
Fig. 1. The profiles of nonlocal kernels: (a) the 1-D projection profile of the nonlocal kernel $J$ defined in (84); (b) the 1-D projection profile of the nonlocal kernel $J$ defined in (85).

Fig. 2. Spinodal decomposition for random initial data with $\phi|_{t=0} = \phi_a + \phi_b \text{rand}(-1, 1)$ where $\phi_a = 0$ and $\phi_b = 0.001$. Snapshots of the phase variable $\phi$ are taken at $t = 10, 20, 30, 50, 100, 200, 500, 600,$ and $1740$.

Table 2
The $L^2$ numerical errors at $t = 0.2$ that are computed by the second-order scheme CN2 using various temporal resolutions. The parameters are listed in (83) and $128^2$ grid points are used to discretize the space.

<table>
<thead>
<tr>
<th>Coarse $\delta t$</th>
<th>Fine $\delta t$</th>
<th>$L^2$-error of $\phi$</th>
<th>Order</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.01</td>
<td>0.005</td>
<td>4.0176e-4</td>
<td></td>
</tr>
<tr>
<td>0.005</td>
<td>0.0025</td>
<td>1.0156e-4</td>
<td>1.983</td>
</tr>
<tr>
<td>0.0025</td>
<td>0.00125</td>
<td>2.5510e-5</td>
<td>1.993</td>
</tr>
<tr>
<td>0.00125</td>
<td>0.000625</td>
<td>6.3907e-6</td>
<td>1.997</td>
</tr>
<tr>
<td>0.000625</td>
<td>0.0003125</td>
<td>1.5992e-6</td>
<td>1.998</td>
</tr>
<tr>
<td>0.0003125</td>
<td>0.00015625</td>
<td>4.0000e-7</td>
<td>1.999</td>
</tr>
</tbody>
</table>

We vary the values of $\phi_a$ and $\phi_b$ to see various coarsening dynamics. In Fig. 2, we perform numerical simulations for initial values with $\phi_a = 0, \phi_b = 0.001$ and order parameters (83). The time step size is $\delta t = 0.001$. We observe the final equilibrium solution that the system from the mixing state to the two phase state. Shortly after the phase separation starts, the domains of the binary components are formed and the interface between different phases can be specified.

Then we keep the same kernel function specified in (84) and set the initial conditions with the randomly perturbed concentration field as follows:

$$\phi(x, y, t = 0) = \phi_a + \phi_b \text{rand}(x, y).$$

(87)
Fig. 3. Spinodal decomposition for random initial data with $\phi|_{t=0} = \phi_a + \phi_b \text{rand}(-1, 1)$ where $\phi_a = 0$ and $\phi_b = 0.01$. Snapshots of the phase variable $\phi$ are taken at $t = 10, 20, 30, 50, 100, 200, 500, 600,$ and $1000$.

Fig. 4. Spinodal decomposition for random initial data with $\phi|_{t=0} = \phi_a + \phi_b \text{rand}(-1, 1)$ where $\phi_a = 0.01$ and $\phi_b = 0.01$. Snapshots of the phase variable $\phi$ are taken at $t = 10, 20, 30, 50, 100, 200, 500, 600,$ and $920$. 
Fig. 5. Time evolution of the free energy functional for the spinodal decomposition with the initial random data of (a) \( \phi_a = 0, \phi_b = 0.001 \), (b) \( \phi_a = 0, \phi_b = 0.01 \), and (c) \( \phi_a = 0.1, \phi_b = 0.01 \). The energy curves show monotone decays for all three cases.

Fig. 6. Nucleation for random initial data with \( \phi |_{t=0} = 0.1 + 0.01 \text{rand}(-1, 1) \) with the kernel \( J \) specified in (85) and \( \gamma_0 = -0.01 \). Snapshots of the phase variable \( \phi \) are taken at \( t = 10, 20, 30, 40, 50, \) and 200.

is obtained after \( t = 1740 \) presents a banded shape, in which the range of \( \phi \) is from \(-0.32\) to \(0.32\). In Fig. 3, when we further change \( \phi_b \) to \(0.01\) and fix \( \phi_a = 0 \), we obtain a totally different equilibrium solution after \( t = 1000 \) that presents a circular shape in which, the value of \( \phi \) in the circle is around \(0.32\). Finally, in Fig. 4, we change \( \phi_a \) to \(0.1\) and keep \( \phi_b = 0.01 \), the obtained equilibrium solution after \( t = 920 \) is still circular shape. However, the value of \( \phi \) in the circle becomes around \(-0.32\). In Fig. 5, we plot the evolution of the energy curves that show the energy monotonically decays with respect to the time for all three cases.

4.3. Nucleation

In this example, we perform numerical simulations of phase nucleation using a kernel function that is not always positive. Use the nonlocal kernel (85) with \( \alpha_1 = 0.1, \alpha_2 = 0.05, \sigma_1 = 0.08 \) and \( \sigma_2 = 0.2 \). Its profile projection in 1D \( (x = y) \) is shown in Fig. 1(b), where one can observe that \( J(x) \) is negative in certain range of \( x \).

We set the initial profile of \( \phi \) as the randomly perturbed concentration field as

\[
\phi(x, y, t = 0) = 0.1 + 0.01 \text{rand}(-1, 1).
\]  

(88)

We vary the parameter \( \gamma_0 \), keeping all other parameters fixed as (83). In Fig. 6, by setting \( \gamma_0 = -0.01 \), snapshots of the phase field variable \( \phi \) are taken at \( t = 10, 20, 30, 40, 50 \) and 200 in which one can observe that after a short time \( (0 \leq t \leq 10) \), the dynamics result in the nucleation of particles (spots). In Fig. 7, we set \( \gamma_0 = 0.1 \) and take the snapshots of the phase field variable \( \phi \) at same time nodes, where different patterns are observed. These two simulations agree well with the numerical results shown in [23,24] qualitatively. In Fig. 8, we plot the evolution of the energy curves for these two cases which shows energy decreasing in time.

5. Concluding remarks

In this paper, we develop two efficient time discretization schemes for solving the nonlocal Cahn–Hilliard equation based on the Invariant Energy Quadratization approach. The proposed schemes possess the advantages of the convex splitting method and the stabilized method, avoiding their deficiency at the same time. More precisely, our schemes are linear, second order accurate and unconditional energy stable. We rigorously proved the well-posedness of the resulted linear system, as well as the unconditional energy stabilities. We remark that, to the best of our knowledge, these schemes are the first linear and second-order-accurate schemes with provable energy stabilities for the nonlocal phase field model. Although we only present proofs for semi-discretized schemes in time in this paper, the results here can be carried over to any consistent finite dimensional Galerkin type approximations since the proofs are all based on a variational formulation with all test functions in the same space as the space of the trial functions.
Fig. 7. Nucleation with random initial data. Here we choose $\phi_{t=0} = 0.1 + 0.01 \text{rand}(-1, 1)$ with the kernel $J$ specified in (85) and $\gamma_0 = 0.1$. Snapshots of the phase variable $\phi$ are taken at $t = 10, 20, 30, 40, 50, 200$.

Fig. 8. Time evolution of the free energy functional for the nucleation examples with (a) $\gamma_0 = -0.01$ and (b) $\gamma_0 = 0.01$. This figure shows energy decays for both cases.

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