Model of interacting equilibrium and non-equilibrium phase transitions

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ABSTRACT
If the system simultaneously undergoes a non-equilibrium phase transition and an equilibrium phase transition accompanied by phase separation, it could be of considerable interest to study their interaction. In this communication the non-equilibrium phase transitions are presented by the canonical chemical models introduced by Schlögl. The equilibrium phase transitions are described on the basis of modified Cahn-Hilliard equation. We consider the advancing fronts which combine these two transformations. Exact solutions are obtained and the conditions of their existence are discussed.

KEYWORDS
phase transition, nonequilibrium phase transition, Cahn-Hilliard equation, Schlögl reactions

1. Introduction

If the system simultaneously undergoes a non-equilibrium phase transition and an equilibrium phase transition accompanied by phase separation, it could be of considerable interest to study their interaction. The canonical models for non-equilibrium phase transitions in chemical reaction systems were introduced by Schlögl [1]. In the non-equilibrium phase transitions the different “phases” correspond to different stationary states of the system. Schlögl considered two reaction systems: the so-called “First Schlögl Reaction”,

\[ A + X \rightleftharpoons 2X, \]  \hspace{1cm} (1.1)

\[ B + X \rightleftharpoons C, \]  \hspace{1cm} (1.2)

and the “Second Schlögl Reaction”,

\[ A + 2X \rightleftharpoons 3X, \]  \hspace{1cm} (1.3)
\[ B + X \rightleftharpoons C. \quad (1.4) \]

The concentrations of species \( A, B \) and \( C \) (which are called the “reservoir reagents”) are presumed to be constant and only concentration of \( X \) can vary with time and space. For the first Schlögl reaction in the absence of diffusion the evolution of \( X \) is described by

\[ \frac{dX}{dt} = -k'_{11}X^2 + k_{11}AX - k_{21}BX + k'_{21}C. \quad (1.5) \]

Here the \( k_{i1}, k'_{i1} \) are the rate constants for the forward and reverse reactions, respectively; the second lower index is “1” for the first Schlögl reaction, and “2” for the second one. Correspondingly, for the second Schlögl reaction in the absence of diffusion the evolution of \( X \) is described by

\[ \frac{dX}{dt} = -k'_{12}X^3 + k_{12}AX^2 - k_{22}BX + k'_{22}C. \quad (1.6) \]

The first reaction exhibits a non-equilibrium phase transition of second order, the second one a phase transition of first order (for the details see [1]).

On the other hand, the commonly accepted phenomenological model for the equilibrium phase transitions is the Cahn-Hilliard equation [2 – 5]. The basic underlying idea of this model is that for inhomogeneous system, e.g. system undergoing a phase transition, the thermodynamic potential (e.g. free energy) should depend not only on the order parameter \( u \), but on its gradient as well. The idea of such dependence was introduced already by Van der Waals in his theory of capillarity [6]. For the inhomogeneous system the local chemical potential \( \mu \) is defined as variational derivative of the thermodynamic potential functional. If the thermodynamic potential is the simplest symmetric (quadratic) function of gradient this leads to the local chemical potential \( \mu \) which depends on Laplacian, or for the one-dimensional case – on the second order derivative of the order parameter. The diffusional flux \( J \) is proportional to the gradient of chemical potential \( \nabla \mu \); the coefficient of proportionality is called mobility \( M \) [7].

With such expression for the flux the diffusion equation instead of usual second order equation becomes a forth-order PDE for the order parameter \( u \) (herein our notations differ from the notations in original papers):

\[ \frac{\partial u}{\partial t'} = \nabla (M \nabla \mu), \quad (1.7) \]

\[ \mu = -\bar{\varepsilon}^2 \Delta u + f(u). \quad (1.8) \]

Here \( M \) is mobility, \( \bar{\varepsilon} \) is usually presumed to be proportional to the capillarity length, and \( f(u) = \frac{d\Phi(u)}{du} \), where \( \Phi(u) \) is homogeneous part of the thermodynamic potential. In the present communication we will take \( f(u) \) in the form of the cubic polynomial (corresponding to the fourth-order polynomial for the homogeneous part of thermodynamic potential):

\[ f(u) = u^3 - \delta u^2 - su. \quad (1.9) \]

Rescaling \( u \) the coefficient at \( u^3 \) could be always scaled to one.
In the present work we consider the model of interacting equilibrium and non-equilibrium phase transitions, which is based on the modified Cahn-Hilliard equation complemented by Schlögl Reactions. We consider the advancing fronts which “combine”, in some sense, these both transitions. To understand the meaning of our modifications, we need to give some insight into the history and existing modifications of CH equation. The classic Cahn-Hilliard equation was introduced as early as in 1958 [2, 3]; the stationary solutions were considered, the linearized version was treated and corresponding instability of homogeneous state identified. However, intensive study of the fully nonlinear form of this equation started essentially later [8]. Now an impressive amount of work is done on nonlinear Cahn-Hilliard equation, as well as on its numerous modifications, see [4, 5]. An important modification was done by Novick-Cohen [9]. Taking into account the dissipation effects which are neglected in the derivation of the classic Cahn-Hilliard equation, she introduced the viscous Cahn-Hilliard (VCH) equation

$$\frac{\partial u}{\partial t} = \nabla \left[ M \nabla \left( \mu + \tilde{\eta} \frac{\partial u}{\partial t} \right) \right],$$  \hspace{1cm} (1.10)

where the coefficient $\tilde{\eta}$ is called viscosity. It was also noticed that VCH equation could be derived as a certain limit of the classic Phase-Field model [10]. Later several authors considered the nonlinear convective Cahn-Hilliard equation (CCH) in one space dimension [11, 13],

$$\frac{\partial u}{\partial t} - \bar{\alpha} u \frac{\partial u}{\partial x} = \frac{\partial}{\partial x} \left[ M \frac{\partial}{\partial x} \left( \mu + \tilde{\eta} \frac{\partial u}{\partial t} \right) \right].$$  \hspace{1cm} (1.11)

Leung [11] proposed this equation as a continual description of lattice gas phase separation under the influence of an external field. Similarly, Emmott and Bray [13] proposed this equation as a model for the spinodal decomposition of a binary alloy in an external field $E$. As they noticed, if the mobility $M$ [7] is independent of the order parameter (concentration), the term involving $E$ will drop out of the dynamics. To get nontrivial results, they presumed the simplest possible symmetric dependence of mobility on the order parameter, viz. $M \sim 1 - ru^2$. Then, they obtained the Burgers-type convection term in equation (1.11) with the coefficient $\bar{\alpha} = 2rE$. Thus, the sign of $\bar{\alpha}$ depends both on the direction of the field and on the sign of $r$. Witelski [12] introduced the equation (1.11) as a generalization of the classic Cahn–Hilliard equation or as a generalization of the Kuramoto–Sivashinsky equation [14, 15] by including a nonlinear diffusion term. In [11, 13] and [16, 17] several approximate solutions and only two exact static kink and anti-kink solutions were obtained. The ‘coarsening’ of domains separated by kinks and anti-kinks was also discussed. To study the joint effects of nonlinear convection and viscosity, Witelski [18] introduced the convective-viscous-Cahn–Hilliard equation (CVCHE) with a general symmetric double-well potential $\Phi(u)$:

$$\frac{\partial u}{\partial t} - \bar{\alpha} u \frac{\partial u}{\partial x} = \frac{\partial}{\partial x} \left[ M \frac{\partial}{\partial x} \left( \mu + \tilde{\eta} \frac{\partial u}{\partial t} \right) \right],$$  \hspace{1cm} (1.12)

$$\mu = -\varepsilon^2 \frac{\partial^2 u}{\partial x^2} + \frac{d\Phi(u)}{du}.$$  \hspace{1cm} (1.13)

It is worth noting that all results, including the stability of solutions, were obtained
without specifying a particular functional form of the potential. Thus, they are valid both for the polynomial and logarithmic potentials. Also, with a constraint imposed on nonlinearity and viscosity, the approximate travelling-wave solutions were obtained. In [19] for equation (1.12) with polynomial potential, see (1.9), and the balance between the applied field and viscosity several exact single- and two-wave solutions were obtained.

Another modification of the nonlinear Cahn-Hilliard equation which attracted much interest is insertion of linear or nonlinear sink/source terms, e.g. due to a chemical reaction, into this equation. Such study was pioneered by Huberman [20]. He introduced Cahn-Hilliard equation with additional kinetic terms corresponding to the reversible first-order autocatalytic chemical reaction and analyzed linear stability of the stationary states. Cohen and Murray [21] considered the same equation in the biological context: they used quadratic nonlinearity to describe growth and dispersal in population model; they studied the stability and identified bifurcations to spatial structures. Apparently unaware of Schlögl paper Huberman [20] and Cohen and Murray [21] in fact considered interplay of equilibrium and (second-order) non-equilibrium phase transitions. Similar equation (with additional nonlinear term) was used in [22] to study segregation dynamics of binary mixtures coupled with chemical reaction. The same equation as in [20][21] was used to describe phase transitions in chemisorbed layer [23], and to model the system of cells that move, proliferate and interact via adhesion [24]. Also, for the latter model several rigorous mathematical results on existence and asymptotics of solutions were obtained [25][26]. General observation is that the presence of chemical reaction can visibly influence the equilibrium phase transition, e.g. freeze the spinodal decomposition or coarsening, stabilizing some stationary inhomogeneous state.

In the present communication we consider the modified Cahn-Hilliard equation complemented by source/sink terms corresponding both to first and second Schlögl reactions. We will call these modifications Cahn-Hilliard-Huberman-Cohen-Murray (CHHCM) and Cahn-Hilliard-Schlögl (CHS) equations respectively. We will also consider the influence of some additional modifications of the Cahn-Hilliard equation, such as viscous and convective terms [9][11][13][18][19].

2. Convective Viscous Cahn-Hilliard-Huberman-Cohen-Murray Equation

In the present Section we first give exact travelling-wave solutions for convective viscous Cahn-Hilliard equation with second order reaction terms. So, we first take into account the influence of both external field and dissipation [9][11][13][18][19]; then we drop the convective and viscous terms, reducing equation to CHHCM equation. To avoid some unnecessary complications we presume reaction (1.2) to be irreversible, i.e. in (1.5) \( k'_{21} = 0 \). In terms of Schlögl model [1] this corresponds to “analog of zero magnetic field” case. From (1.12), (1.8), (1.9) and (1.5) we write down the Convective Viscous CHHCM equation, first in terms of the initial variable \( X \) (concentration):

\[
\frac{\partial X}{\partial t'} - \hat{\alpha} X \frac{\partial^2 X}{\partial x'^2} = M \frac{\partial}{\partial x'^2} \left( \hat{\mu} + \hat{\eta} \frac{\partial X}{\partial t'} \right) - k'_{11} X^2 + k_{11} A X - k_{21} B X,
\]

where

\[
\hat{\mu} = -\bar{\varepsilon}^2 \frac{\partial^2 X}{\partial x'^2} + \bar{f} (X),
\]

(2.1, 2.2)
\[ f(X) = qX^3 - \delta X^2 - sX. \] (2.3)

The equations (2.1)-(2.3) presume implicitly that in the system \( A - B - C - X \) the components \( A \) and \( B \) are in large excess, are not essentially exhausted during the chemical reaction and not essentially changed due to phase transition; we have also presumed \( M \) to be a constant. We renormalize \( X, x' \) and \( t' \):

\[ X = uX_0; \quad x' = xl; \quad t' = t\tau. \] (2.4)

Here \( X_0 = \frac{1}{\sqrt{q}}, \quad \tau = \frac{1}{k_{11}X_0} = \frac{1}{\sqrt{q}} \) and \( l = \sqrt{M\tau} = \frac{M}{k_{11}} \). Denoting \( \alpha = \bar{\alpha}X_0/\tau = \frac{\bar{\alpha}}{\sqrt{q}} \), \( \varepsilon^2 = \frac{\varepsilon^2}{\tau}, \quad \eta = \frac{\eta}{\tau}, \quad \delta = \frac{\delta}{X_0} = \frac{\tilde{\delta}}{\sqrt{q}} \) and \( s = \tilde{s}q \) we write down equation (2.1) in the non-dimensional form

\[ \frac{\partial u}{\partial t} - \alpha u \frac{\partial u}{\partial x} = \frac{\partial^2}{\partial x^2} \left( -\varepsilon^2 \frac{\partial^2 u}{\partial x^2} + u^3 - \delta u^2 - su + \eta \frac{du}{dt} \right) - u(u - u_1). \] (2.5)

We have also introduced

\[ u_1 = \frac{k_{11}A - k_{21}B}{k'_{11}X_0}, \] (2.6)

presuming \( u_1 > 0 \), i.e. \( k_{11}A > k_{21}B \). Looking for the travelling wave solutions of (2.5) we introduce the travelling wave coordinate \( z = x - vt \). This yields

\[ \frac{d}{dz} \left[ vu + \frac{\alpha u^2}{2} + \frac{d}{dz} \left( -\varepsilon^2 \frac{d^2 u}{dz^2} + u^3 - \delta u^2 - su - v\eta \frac{du}{dz} \right) \right] = u(u - u_1). \] (2.7)

We look for the solution, which connects the stationary state of the reaction system \( u = u_1 \) at \( z = -\infty \) with the stationary state \( u = 0 \) at \( z = +\infty \). The simplest possible Ansatz for the anti-kink solution (as usually we call “kinks” the solutions with \( \frac{du}{dz} > 0 \), and “anti-kinks” - solutions with \( \frac{du}{dz} < 0 \) with this property will be

\[ \frac{du}{dz} = \kappa u (u - u_1), \] (2.8)

where \( \kappa \) is presently unknown positive constant. Then equation (2.7) could be written as

\[ \frac{d}{dz} \left[ vu + \frac{\alpha u^2}{2} - \frac{1}{\kappa} u + \frac{d}{dz} \left( -\varepsilon^2 \frac{d^2 u}{dz^2} + u^3 - \delta u^2 - su - v\eta \frac{du}{dz} \right) \right] = 0. \] (2.9)

Integrating once, we get

\[ vu + \frac{\alpha u^2}{2} - \frac{1}{\kappa} u + \frac{d}{dz} \left( -\varepsilon^2 \frac{d^2 u}{dz^2} + u^3 - \delta u^2 - su - v\eta \frac{du}{dz} \right) = C_1. \] (2.10)

Regarding the Ansatz (2.8), for the latter equation to be satisfied the expression under the derivative should be proportional to \( u \). I.e., for (2.8) to give the solution of (2.5),
two equations should be satisfied,

\[ vu + \frac{u^2}{2} - \frac{1}{\kappa} u + \beta \frac{du}{dz} = C_1, \]  

(2.11)

\[ - \varepsilon^2 \frac{d^2 u}{dz^2} + u^3 - \delta u^2 - su - v\eta \frac{du}{dz} = \beta u + C_2, \]  

(2.12)

where \( \beta, C_1 \) and \( C_2 \) are constants. The expression for the second derivative of \( u \) is easily written as:

\[ \frac{d^2 u}{dz^2} = \kappa^2 (2u^3 - 3u_1u^2 + u_1^2 u). \]  

(2.13)

Then Eqs. (2.11), (2.12) take the form

\[ \left( \frac{\alpha}{2} + \beta \kappa \right) u^2 + \left( v - \frac{1}{\kappa} - \beta \kappa u_1 \right) u = C_1, \]  

(2.14)

\[ - \varepsilon^2 \kappa^2 \left[ 2u^3 - 3u_1u^2 + u_1^2 u \right] + u^3 - \delta u^2 - (s + \beta) u - v\eta \kappa (u^2 - u_1 u) = C_2. \]  

(2.15)

These equations should be satisfied for arbitrary \( u \). Rearranging the terms and equating coefficients at each power of \( u \) to zero we finally obtain five constraints on the parameters:

\[ \frac{\alpha}{2} + \beta \kappa = 0, \]  

(2.16)

\[ v = \frac{1}{\kappa} + \beta \kappa u_1, \]  

(2.17)

\[ \kappa^2 = \frac{1}{2\varepsilon^2}, \]  

(2.18)

\[ v\eta \kappa = \frac{3}{2} u_1 - \delta, \]  

(2.19)

\[ v\eta \kappa u_1 = \frac{1}{2} u_1^2 + s + \beta. \]  

(2.20)

There are five constraints (2.16)-(2.20) and only three unknown variables \( \kappa, v \) and \( \beta \). I.e. for the constant velocity transition front to exist, two additional constraints on the values of the stationary states of the reaction system and on the values of the equilibrium states for the phase transition should be imposed. Now, there is some freedom in selecting which parameters are “basic”, those related to reaction system, or
those related to the “Cahn-Hilliard part”. Presuming the latter to be basic we write down the constraints as

\[ u_1 = \frac{2\sqrt{2\varepsilon}(\delta + \eta)}{3\sqrt{2\varepsilon} + \alpha \eta}, \]  

(2.21)

\[ s = \left[ \frac{2\sqrt{2\varepsilon}(\delta + \eta)}{3\sqrt{2\varepsilon} + \alpha \eta} \right]^2 - \frac{2\sqrt{2\varepsilon}(\delta + \eta)}{3\sqrt{2\varepsilon} + \alpha \eta} + \frac{\alpha \varepsilon}{\sqrt{2}}. \]  

(2.22)

If the constraints (2.16)-(2.20) are satisfied, the solution of equation (2.8) is simultaneously solution of the travelling-wave equation (2.7). Integrating (2.8) once, we get

\[ u = \frac{u_1 \exp \{-\kappa u_1 (z + \phi)\}}{1 + \exp \{-\kappa u_1 (z + \phi)\}}, \]  

(2.23)

where \( \phi \) is an arbitrary constant. It is natural to take position of the maximal value of the derivative \( \frac{du}{dz} \) (when \( \frac{d^2u}{dz^2} = 0 \)) as \( z = 0 \); then \( \phi = 0 \). The solution (2.23) could be rewritten in the form

\[ u = \frac{u_1}{2} \left[ 1 - \tanh \left( \frac{u_1}{2\sqrt{2\varepsilon}}(x - vt) \right) \right]. \]  

(2.24)

Here we have used \( \kappa = \frac{1}{\sqrt{2\varepsilon}} \); see (2.18); the velocity \( v \) of the transition front is given by (2.16) and (2.17),

\[ v = \sqrt{2\varepsilon} + \beta \kappa u_1 = \sqrt{2\varepsilon} - \frac{1}{2}\alpha u_1. \]  

(2.25)

The roots of equation

\[ \tilde{u} \left( \tilde{u}^2 - \delta \tilde{u} - s \right) = 0. \]  

(2.26)

correspond to the extrema of the homogeneous part of the thermodynamic potential (1.8), (1.9), \( \tilde{u}_1, \tilde{u}_3 \) to stable minima and \( \tilde{u}_2 \) to unstable maximum. The root \( \tilde{u}_3 = 0 \) coincides with one of the stationary states of the reaction system. Substitution of (2.22) into the latter equation for \( s \) yields two remaining roots, i.e. two constraints imposed on the values of \( \tilde{u}_1, \tilde{u}_2 \) and \( u_1 \),

\[ \tilde{u}_{1,2} = \frac{1}{2} \left[ \delta \pm \sqrt{G} \right], \]  

(2.27)

\[ G = \delta^2 + 4 \left\{ \left[ \frac{2\sqrt{2\varepsilon}(\delta + \eta)}{3\sqrt{2\varepsilon} + \alpha \eta} \right]^2 - \frac{2\sqrt{2\varepsilon}(\delta + \eta)}{3\sqrt{2\varepsilon} + \alpha \eta} + \frac{\alpha \varepsilon}{\sqrt{2}} \right\}. \]  

(2.28)

Here the discriminant of quadratic equation is denoted by \( G \) for convenience. To understand the mutual influence of the equilibrium and non-equilibrium transitions it is practical to consider several special cases of (2.27)-(2.28). First we consider the CHHCM case, i.e. the absence of the applied field and dissipation. For \( \alpha = 0 \) and
\( \eta = 0 \) expression (2.28) simplifies drastically, yielding \( G = \frac{1}{9} \delta^2 \). Then (2.21) and (2.27) become

\[
\dot{u}_1 = \frac{2\delta}{3}; \quad \ddot{u}_{1,2} = \frac{1}{2} \left[ \delta \pm \frac{\delta}{3} \right]. \quad (2.29)
\]

This means, that for the constant-velocity transition front to exist, the values of the order parameter corresponding to the stationary states of the chemical reactions system, \( u_1, 0 \), should coincide exactly with the values, corresponding to the equilibrium phases, i.e. \( u_1 = \ddot{u}_1; u_3 = 0 \). The thermodynamic potential should be symmetric, \( \ddot{u}_2 = \frac{u_1}{2} \), with equal-depth wells. The velocity depends on the \( \varepsilon \) only, \( v = \sqrt{2} \varepsilon \). Now let \( \alpha = 0; \eta \neq 0 \). From (2.21), (2.27) and (2.28) we obtain

\[
u_1 = \frac{2(\delta + \eta)}{3}; \quad \ddot{u}_{1,2} = \frac{1}{2} \left[ \delta \pm \frac{\delta + 4\eta}{3} \right]. \quad (2.30)
\]

I.e., the stable stationary value for the reaction system should again coincide with the stable stationary value for the equilibrium transition, but the unstable value for the equilibrium transition should be shifted to the lower value. As it was mentioned in the introduction, the derivative of the homogeneous part of the thermodynamic potential \( \Phi(u) \) is given by (1.9):

\[
\frac{d\Phi(u)}{du} = u^3 - \delta u^2 - su. \quad (2.31)
\]

Integrating once and substituting value of \( \ddot{u}_1 \) and \( s \) for \( \alpha = 0 \) we obtain the following expressions for the potential values \( \Phi(\ddot{u}_1) \) and \( \Phi(\ddot{u}_3) \),

\[
\Phi(\ddot{u}_1) = -\frac{\eta}{6} \left( \frac{2(\delta + \eta)}{3} \right)^3 + C; \quad \Phi(\ddot{u}_3) = \Phi(0) = C. \quad (2.32)
\]

That is, to compensate the dissipation, the potential well corresponding to \( \ddot{u}_1 \) should be deeper.

On the other hand, if \( \alpha \neq 0; \eta = 0 \) we get

\[
u_1 = \frac{2\delta}{3}; \quad \ddot{u}_{1,2} = \frac{\delta}{2} \left[ 1 \pm \frac{1}{3} \sqrt{1 + 108 \frac{\alpha \varepsilon}{\sqrt{2} \delta^2}} \right]. \quad (2.33)
\]

This means, that for positive \( \alpha \) the order parameter value for the final state after transition, \( u = u_1 \), is somewhat lower than the equilibrium value \( \ddot{u}_1 \); the unstable equilibrium value \( \ddot{u}_2 \) should be somewhat lower too.

Now let both \( \alpha \neq 0, \eta \neq 0 \). The expression for the velocity (2.25) is independent on \( \eta \); for the special value \( \alpha = \frac{3\sqrt{2} \varepsilon}{u_1} \) (and \( \eta \neq 0 \)) the velocity is zero, i.e. for the corresponding value of the applied field the transition front becomes static. Using (2.21) we get \( \alpha = \frac{3\sqrt{2} \varepsilon}{u_1} \). Substitution of this value of \( \alpha \) into (2.21), (2.27) and (2.28) yields

\[
\ddot{u}_{1,2} = \frac{\delta}{2} \left( 1 \pm \frac{1}{3} \sqrt{1 + 108 \frac{\varepsilon^2}{\delta^3}} \right). \quad (2.34)
\]
Interestingly, the viscosity $\eta$ has dropped out from the latter expression. It is physically reasonable: there is no dissipation for the static transition front; the deviation of the order parameter value $u = u_1$ for the final state after transition from its equilibrium value $\bar{u}_1$ is exactly the same, as given by (2.33) (i.e. for $\eta = 0$ case) for this special value of $\alpha$.

### 3. Convective Viscous Cahn-Hilliard-Schlögl Equation

In this Section we first give exact travelling-wave solutions for convective viscous Cahn-Hilliard equation with third order reaction terms. Again, we first take into account the influence of both external field and dissipation [9, 11–13, 18, 19]; then we drop the convective and viscous terms, reducing equation to CHS equation. To make the calculations somewhat more transparent we presume reaction (1.4) to be irreversible, i.e. in (1.6) $k_2' = 0$. From (1.12), (1.8), (1.9) and (1.6) we write down the Convective Viscous CHS equation, first in terms of the initial variable $X$ (concentration):

$$
\frac{\partial X}{\partial t'} - \bar{\alpha} X \frac{\partial X}{\partial x'} = M \frac{\partial^2 X}{\partial x'^2} \left( \bar{\mu} + \bar{\eta} \frac{\partial X}{\partial t'} \right) - k_{12} X^3 + k_{12} A X^2 - k_{22} B X ,
$$

(3.1)

$$
\bar{\mu} = -\bar{\varepsilon}^2 \frac{\partial^2 X}{\partial x'^2} + \bar{f} (X),
$$

(3.2)

$$
\bar{f} (X) = q X^3 - \bar{\delta} X^2 - \bar{s} X.
$$

(3.3)

Writing down equations (3.1)-(3.3) we again presume implicitly that in the system $A - B - C - X$ the components $A$ and $B$ are in large excess and are not essentially exhausted during the chemical reaction; we have also presumed $M$ to be a constant.

We renormalize $X$, $x'$ and $t'$,

$$X = uX_0; \; x' = xl; \; t' = t\tau.
$$

(3.4)

Here $X_0 = \frac{1}{\sqrt{q}}$, $\tau = \frac{1}{\sqrt{k_{12}A_0}} = \frac{q}{k_{12}}$, and $l = \sqrt{M\tau} = \sqrt{M} = \sqrt{Mq}$. Denoting $\alpha = \bar{\alpha} \frac{X_0}{\sqrt{k_{12}M}}$, $\varepsilon^2 = \bar{\varepsilon}^2 \tau$, $\eta = \frac{\bar{\eta}}{\tau}$, $\delta = \frac{\bar{\delta}}{X_0} = \bar{\delta} \sqrt{q}$; $s = \bar{s}q$; $R = \frac{k_{12}A}{k_{12}A_0}$ and $Q = \frac{k_{22}B}{k_{12}A_0}$, we write down equation (3.1) in non-dimensional form

$$
\frac{\partial u}{\partial t} - \alpha u \frac{\partial u}{\partial x} = \frac{\partial^2}{\partial x^2} \left( -\varepsilon^2 \frac{\partial^2 u}{\partial x^2} + u^3 - \delta u^2 - su + \eta \frac{\partial u}{\partial t} \right) - u \left( u^2 - Ru + Q \right). \tag{3.5}
$$

Below we presume that the quadratic equation

$$u^2 - Ru + Q = 0 \tag{3.6}
$$

always has real roots $u_1, u_2$, $u_1 \geq u_2$; i.e. $R^2 - 4Q \geq 0$ which means, in terms of the parameters of the reaction system, $(k_{12}A)^2 \geq 4k_{12}'k_{22}B$. Looking for the travelling
wave solutions of (3.5) we introduce the travelling wave coordinate \( z = x - vt \). This yields

\[
\frac{d}{dz} \left[ vu + \frac{u^2}{2} + \frac{d}{dz} \left( -\varepsilon^2 \frac{d^2 u}{dz^2} + u^3 - \delta u^2 - su - v\eta \frac{du}{dz} \right) \right] = u(u - u_1)(u - u_2).
\]

(3.7)

As in the previous Section, we look for the solution, which connects the stationary state of the reaction system \( u = u_1 \) at \( z = -\infty \) with the stationary state \( u = 0 \) at \( z = +\infty \). So, the proper Ansatz for the anti-kink solution will be again (2.8)

\[
\frac{1}{\kappa} \frac{du}{dz} = u(u - u_1),
\]

(3.8)

where \( \kappa \) is presently unknown positive constant. Then equation (3.7) could be written as

\[
\frac{d}{dz} \left[ vu + \frac{u^2}{2} + \frac{d}{dz} \left( -\varepsilon^2 \frac{d^2 u}{dz^2} + u^3 - \delta u^2 - su - v\eta \frac{du}{dz} \right) \right] = \frac{d}{dz} \left( \frac{1}{2\kappa} u^2 - \frac{u_2}{\kappa} u \right).
\]

(3.9)

Integrating once, we get

\[
\left( v + \frac{u_2}{\kappa} \right) u + \left( \alpha - \frac{1}{\kappa} \right) \frac{u^2}{2} + \frac{d}{dz} \left( -\varepsilon^2 \frac{d^2 u}{dz^2} + u^3 - \delta u^2 - su - v\eta \frac{du}{dz} \right) = C_1.
\]

(3.10)

Regarding the Ansatz (2.8), for the latter equation to be satisfied the expression under the derivative should be proportional to \( u \). I.e., for (3.8) to give the solution of (3.7) two equations should be satisfied,

\[
\left( v + \frac{u_2}{\kappa} \right) u + \left( \alpha - \frac{1}{\kappa} \right) \frac{u^2}{2} + \beta \frac{du}{dz} = C_1,
\]

(3.11)

\[
-\varepsilon^2 \frac{d^2 u}{dz^2} + u^3 - \delta u^2 - su - v\eta \frac{du}{dz} = \beta u + C_2,
\]

(3.12)

where \( C_1, C_2 \) and \( \beta \) are constants. The expression for the second derivative of \( u \) is given again by Eq. (2.13). Then Eqs. (3.11), (3.12) take the form

\[
\left( v + \frac{u_2}{\kappa} \right) u + \left( \alpha - \frac{1}{\kappa} \right) \frac{u^2}{2} + \beta \kappa (u^2 - u_1 u) = C_1,
\]

(3.13)

\[
-\varepsilon^2 \kappa^2 \left( 2u^3 - 3u_1 u^2 + u_1^2 u \right) + u^3 - \delta u^2 - (s + \beta) u - v\eta \kappa (u^2 - u_1 u) = C_2.
\]

(3.14)

These equations should be satisfied for arbitrary \( u \). Rearranging the terms and equating coefficients at each power of \( u \) to zero we finally obtain five constraints on the parameters:

\[
2\varepsilon^2 \kappa^2 = 1; \quad \kappa^2 = \frac{1}{2\varepsilon^2}; \quad \kappa = \frac{1}{\sqrt{2\varepsilon}},
\]

(3.15)
\[ \frac{3}{2} u_1 - \delta - v \eta \kappa = 0, \quad (3.16) \]

\[ - \frac{1}{2} u_1^2 - s - \beta + v \eta \kappa u_1 = 0, \quad (3.17) \]

\[ \beta = \frac{1}{2 \kappa^2} - \frac{\alpha}{2 \kappa}, \quad (3.18) \]

\[ v = \beta \kappa u_1 - \frac{u_2}{\kappa}. \quad (3.19) \]

Similarly to (2.16)-(2.20) there are five constraints (3.15)-(3.19) and only three unknowns \( \kappa \), \( v \) and \( \beta \). I.e. for the constant velocity transition front to exist, two additional constraints on the values of the stationary states of the reaction system and on the values of the equilibrium states for the phase transition should be imposed. Again, there is freedom in selecting which parameters are “basic”, those related to reaction system, or those related to the “Cahn-Hilliard part”. However, here it is more convenient to presume the former to be basic. The constraints are

\[ \delta = \frac{3}{2} u_1 - \left( \frac{u_1}{2} - u_2 \right) \eta + \frac{\alpha u_1}{2 \sqrt{2 \varepsilon}} \eta; \quad (3.20) \]

\[ s = -\frac{1}{2} u_1^2 - \varepsilon^2 + \left( \frac{u_1}{2} - u_2 \right) \eta + \frac{\alpha}{\sqrt{2}} \left( \varepsilon - \frac{u_2}{2 \varepsilon} \right). \quad (3.21) \]

The latter expressions impose evident limitations on the roots of

\[ \tilde{u} \left( u^2 - \delta \tilde{u} - s \right) = 0, \quad (3.22) \]

i.e. on the extrema of the homogeneous part of the thermodynamic potential, here \( \tilde{u}_1 \), \( \tilde{u}_3 \) correspond to stable minima and \( \tilde{u}_2 \) to unstable maximum. The root \( \tilde{u}_3 = 0 \) coincides with one of the stationary states of the reaction system. The expressions for two remaining roots yield two constraints imposed on the values of \( \tilde{u}_1 \), \( \tilde{u}_2 \) and \( u_1 \). The velocity of the transition front \( v \) is

\[ v = \sqrt{2 \varepsilon} \left( \frac{u_1}{2} - u_2 \right) - \frac{\alpha u_1}{2}. \quad (3.23) \]

To understand the mutual influence of the equilibrium and non-equilibrium transitions it is again expedient to consider several special cases of (3.20) and (3.21). First we consider the CHS case, i.e. the absence of the applied field and dissipation. For \( \alpha = 0 \) and \( \eta = 0 \) these expressions simplify drastically, yielding

\[ \delta = \frac{3}{2} u_1; \quad s = -\frac{1}{2} u_1^2 - \varepsilon^2 \quad (3.24) \]
and, correspondingly

\[ \tilde{u}_{1,2} = \frac{u_1}{4} \left( 3 \pm \sqrt{1 - \frac{16 \varepsilon^2}{u_1^2}} \right) \approx \frac{u_1}{4} \left[ 3 \pm \left( 1 - 8 \frac{\varepsilon^2}{u_1^2} \right) \right]. \quad (3.25) \]

I.e., even in the absence of the applied field and viscosity the order parameter value for the final state after transition, \( u = u_1 \), is somewhat higher than the equilibrium value \( \tilde{u}_1 \). The velocity is

\[ v = \sqrt{2 \varepsilon} \left( \frac{u_1}{2} - u_2 \right). \quad (3.26) \]

Remarkably, the dependence of velocity on the stationary values of concentration, \( u_1, u_2, 0 \), is exactly the same as for the well known travelling-wave solution for the diffusion equation with cubic nonlinearity; for \( u_2 = \frac{u_1}{2} \) the velocity is zero, that is the front becomes static. However, the coefficient in (3.26) depends on \( \varepsilon \), i.e. on the “Cahn-Hilliard part”. As it was mentioned in the previous Section, the derivative of the homogeneous part of the thermodynamic potential \( \Phi (u) \) is given by (2.31). Integrating once and substituting values of \( \delta \) and \( s \) given by (3.24) we obtain the following expression for the potential \( \Phi (u) \)

\[ \Phi (u) = \frac{1}{4} u^4 - \frac{1}{2} u_1 u^3 + \frac{1}{2} \left( \frac{1}{2} u_1^2 + \varepsilon^2 \right) u^2 + C, \quad (3.27) \]

where \( C \) is a constant. Then final (after transition) value of the potential is \( \Phi (u_1) = \frac{1}{2} \varepsilon^2 u_1^2 + C \). Taking into account \( \varepsilon \ll 1 \), to calculate the equilibrium value \( \Phi (\tilde{u}_1) \) we use the approximate expression from (3.25), \( \tilde{u}_1 \approx u_1 - 2 \frac{\varepsilon^2}{u_1^2} \). Substitution into (3.27) and neglecting higher order in \( \varepsilon^2 \) terms yields \( \Phi (\tilde{u}_1) \approx \frac{1}{2} \varepsilon^2 u_1^2 + C \), i.e. it is nearly equal to the value after transition.

It means, that despite the deviation of the concentration in the final state after transition from its equilibrium value, the deviations of thermodynamic potential from its equilibrium value are of the higher order in \( \varepsilon^2 \). Now let \( \alpha = 0; \ \eta \neq 0 \) in (3.20), (3.21),

\[ \tilde{u}_{1,2} = \frac{1}{2} \left\{ \frac{3}{2} u_1 - \left( \frac{u_1}{2} - u_2 \right) \eta \pm \sqrt{\left[ \frac{u_1}{2} + \left( \frac{u_1}{2} - u_2 \right) \eta \right]^2 - 4 \varepsilon^2} \right\}. \quad (3.28) \]

From (3.20) \( \left( \frac{u_1}{2} - u_2 \right) = \frac{v}{\sqrt{2 \varepsilon}} \); comparing (3.25) and (3.28) we see that the deviation term has the form \( \frac{v \eta}{\sqrt{2 \varepsilon}} \), i.e. multiple of velocity and viscosity. On the other hand, if \( \alpha \neq 0; \ \eta = 0 \),

\[ \tilde{u}_{1,2} = \frac{u_1}{4} \left[ 3 \pm \sqrt{1 + \frac{16 \varepsilon}{u_1^2} \left( \frac{\alpha}{\sqrt{2} \varepsilon} - \varepsilon \right)} \right]. \quad (3.29) \]

Now, let both \( \alpha \) and \( \eta \) be non-zero. The expression for the velocity (3.23) is inde-
dependent on $\eta$; for the special value of $\alpha$,

$$\alpha = \frac{2\sqrt{2\varepsilon}}{u_1} \left( \frac{u_1}{2} - u_2 \right),$$  \hspace{1cm} (3.30)

the velocity is zero, i.e. for the corresponding value of the applied field the transition front becomes static even for $u_2 \neq \frac{u_1}{2}$. Substitution of this value of $\alpha$ into (3.20) and (3.21) yields

$$s = -\frac{1}{2} u_1^2 - \frac{2\varepsilon^2 u_2}{u_1}; \quad \delta = \frac{3}{2} u_1,$$  \hspace{1cm} (3.31)

and

$$\tilde{u}_{1,2} = \frac{1}{4} u_1 \left( 3 \pm \sqrt{1 - 32\varepsilon^2 u_2 u_1^3} \right).$$  \hspace{1cm} (3.32)

Again the viscosity $\eta$ has self-consistently dropped out from the latter expression, there is no dissipation for the static transition front; the deviation of the order parameter value $u_1$ after transition from its equilibrium value $\tilde{u}_1$ is exactly the same, as given by (3.29) (i.e. for $\eta = 0$ case) for this special value of $\alpha$.

4. Discussion

In the present work we have modeled the interplay of equilibrium and non-equilibrium phase transitions. The equilibrium phase transitions are described on the basis of modified Cahn-Hilliard equation [18,19]. The non-equilibrium phase transitions are presented by the canonical chemical models introduced by Schlögl [1]. In these models the different “phases” correspond to different stationary states of the chemical reactions system. Schlögl considered two reaction systems: the so-called “First Schlögl Reaction” (1.1)-(1.2), which is analog of the second order equilibrium phase transition, and the “Second Schlögl Reaction” (1.3)-(1.4), which is analog of the first order equilibrium phase transition, for details see [1]. Each of these reaction systems has four components, however the concentrations of three reagents (so called “reservoir reagents”) are assumed to be kept constant, and only one reagent’s concentration changes in time and space. If the system is well mixed (or there is no spacial mass transfer) the time evolution of this reagent is governed by a nonlinear ordinary differential equation. It is quadratic polynomial nonlinearity for the First Schlögl Reaction (1.5), and the cubic nonlinearity for the Second Schlögl Reaction (1.6). If the mass transfer should be taken into account, it is usually described by diffusion equation. However, if the system is essentially inhomogeneous, e.g. undergoes a phase transition, the proper description of the mass transfer is given by the Cahn-Hilliard equation [2–5], complemented with nonlinear sink/source terms. For the second-order reaction system such approach was pioneered by Huberman [20] and Cohen and Murray [21]. Apparently unaware of Schlögl paper they in fact considered interplay of equilibrium and (second-order) non-equilibrium phase transitions. Huberman introduced Cahn-Hilliard equation with additional kinetic terms corresponding to the reversible first-order autocatalytic chemical reaction. He analyzed linear stability of the stationary states and mutual influence of spinodal decomposition and reaction. Cohen and Murray considered the same equation in the
biological context; using the nonlinear stability analysis based on a multi-scale perturbation method they identified bifurcations to spatial structures. The same equation as in [20,21] was used to describe phase transitions in chemisorbed layer [23] and to model the system of cells that move, proliferate and interact via adhesion [24]. On the other hand, to the best of our knowledge there is no study of the Cahn-Hilliard equation with third order reaction term in the literature.

Our aim in the present work was to consider the possibly simple situation, where the interplay of the equilibrium and non-equilibrium phase transitions could be observed explicitly. So, we considered the advancing fronts which “combine”, in some sense, these both transitions. We obtained several exact travelling wave solutions, which exhibit explicit parametric dependence. Naturally, for both transitions to proceed simultaneously some additional constraints should be imposed on the parameters of the model.

To get more direct insight here we return to dimensional parameters. Starting from the CHHCM equation supplemented by additional convective term and viscosity we see that the coexistence of equilibrium and non-equilibrium transformations in the form of constant-velocity transition front imposes quite rigid constraints on the parameters. From (2.25) the dimensional velocity \( V = \frac{\nu l}{\tau} \) is

\[
V = k'_{11} \frac{\sqrt{2}{\bar{\alpha}X_1\sqrt{q}}}{k'_{11}} = \sqrt{2}k'_{11}\bar{\varepsilon} - \bar{\alpha}X_1 .
\]  

(4.1)

Here \( X_1 = u_1X_0 \) is the dimensional stationary concentration of the reaction system; from (2.6) it follows

\[
X_1 = u_1X_0 = \frac{k_{11}A - k_{21}B}{k'_{11}} .
\]

(4.2)

Remarkably, in the absence of the field, \( \bar{\alpha} = 0 \), the velocity does not depend on this concentration, but on the parameters of the “Cahn-Hilliard part” \( q, \varepsilon \) and on the reaction constant for the reverse first reaction (1.1) only,

\[
V = \sqrt{2}k'_{11}\bar{\varepsilon} .
\]

(4.3)

In this case the velocity of the anti-kink solution is always positive, and of the kink-solution negative. I.e. the stable state \( X_1 \) of the chemical system always spreads on the cost of unstable zero state. In the absence of the field and viscosity the constraints imposed on the stationary states of the chemical system and on the stationary values of polynomial part of the chemical potential \( \tilde{X}_i = \tilde{u}_iX_0 = \tilde{u}_i/\sqrt{q} \) are very rigid indeed

\[
X_1 = \tilde{X}_1 ; \ X_3 = \tilde{X}_3 = 0 ; \ \tilde{X}_2 = \frac{1}{2}\tilde{X}_1 ;
\]

(4.4)

i.e. the stationary states for the reaction system should coincide with the stationary states for equilibrium transition. This means also that the homogeneous part of the thermodynamic potential \( \Phi \) should be a symmetric function with equal-depth wells. Now, if the viscosity is non-zero (but still \( \bar{\alpha} = 0 \)) the expression for the velocity (4.3) is not changed; the exact expressions for the stationary concentrations of the reactions
system still are (see (2.30)):

\[ X_1 = \tilde{X}_1; \ X_3 = \tilde{X}_3 = 0. \] (4.5)

I.e., the stationary states for the reactions system should again coincide with the stable states for equilibrium transition; however the unstable state \( \tilde{X}_2 \) should be shifted to the lower value,

\[ \tilde{X}_2 = \frac{1}{2} \tilde{X}_1 - \frac{\bar{\eta} k'_{11}}{q}. \] (4.6)

So, to compensate the additional dissipation the homogeneous part of the thermodynamic potential \( \Phi \) becomes asymmetric, the potential well corresponding to \( X_1 \) is now deeper, see (2.32); the difference, naturally, disappears for zero viscosity \( \bar{\eta} \). On the other hand, if \( \bar{\alpha} \neq 0; \ \bar{\eta} = 0 \),

\[ X_1 = \frac{2 \delta}{3}; \ \tilde{X}_{1,2} = \frac{\delta}{2} \left[ 1 \pm \frac{1}{3} \sqrt{1 + 36 \frac{\bar{\alpha} \bar{\varepsilon}}{2 M \delta^2 q^2}} \right]. \] (4.7)

I.e., for positive \( \bar{\alpha} \) the order parameter value for the final state after transition, \( X = X_1 \), is somewhat lower than the equilibrium value \( \tilde{X}_1 \); so the presence of the field can prevent final equilibration. The unstable equilibrium value \( \tilde{X}_2 \) should be somewhat lower too, so the potential \( \Phi \) is again asymmetric. The correction depends only on the “Cahn-Hilliard part parameters”, including the mobility \( M \). If both \( \alpha \neq 0, \ \eta \neq 0 \), for the special value of the applied field,

\[ \bar{\alpha} = \frac{3 \sqrt{2} \bar{\varepsilon}}{\delta \sqrt{q}} k'_{11}, \] (4.8)

the velocity is zero, i.e. the transition front becomes static. The latter expression depends both on the Cahn-Hilliard parameters and on \( k'_{11} \), so the static front is due to the balance of equilibrium and reactive processes. The viscosity \( \eta \) has dropped out from the corrections to the stationary states. It is physically reasonable: there is no dissipation for the static transition front; the deviation of the order parameter value \( X = X_1 \) for the final state after transition from its equilibrium value \( \tilde{X}_1 \), see (2.34), is exactly the same as given by (4.7) (i.e. for \( \bar{\eta} = 0 \) case) for this special value of \( \bar{\alpha} \).

Now, considering convective viscous CHS equation (3.1), we return to dimensional parameters again. From (3.26) the dimensional velocity \( V = v l/\tau \) is now

\[ V = \sqrt{2} \frac{k'_{12}}{\sqrt{q}} \bar{\varepsilon} \left( \frac{X_1}{2} - X_2 \right) - \frac{\bar{\alpha} X_1}{2}. \] (4.9)

As compared to the second order non-equilibrium phase transition, the situation for the first order non-equilibrium phase transition is much more "flexible". As in Section 3, we consider first the absence of the field, \( \bar{\alpha} = 0 \):

\[ V = \sqrt{2} \frac{k'_{12}}{\sqrt{q}} \bar{\varepsilon} \left( \frac{X_1}{2} - X_2 \right). \] (4.10)
Comparing the latter equation with (4.3) we see that this expression is very similar to the coefficient in (4.10) (to avoid confusion we remind that $k'_1$ and $k'_2$ have different dimensionality). However, the dependence of velocity on the stationary values of concentration, $X_1$, $X_2$, 0, is exactly the same as for the well known travelling-wave solution for the diffusion equation with cubic nonlinearity; for $X_2 = \frac{X_1}{2}$ the velocity is zero, that is the front becomes static. Also, for zero field the viscosity $\eta$ enters the constraints (3.20) and (3.21) always multiplied by $(\frac{X_1}{2} - X_2)$, see (3.28). Particularly, for the static front the stationary concentrations $\bar{X}_1$, $\bar{X}_2$ will not depend on $\bar{\eta}$, which is reasonable physically. If additionally to $\bar{\alpha} = 0$ it is also $\bar{\eta} = 0$, that is the CHS-case, the final value after transition $X_1$ will deviate from the equilibrium value, see (3.25). Taking into account $\bar{\varepsilon} \ll 1$, we get

$$X_1 \simeq \bar{X}_1 + 2 \frac{\bar{\varepsilon}^2 k'_2}{X_1 M q^2}.$$  (4.11)

I.e., even in the absence of the applied field and viscosity the order parameter value for the final state after transition, $X = X_1$, is somewhat higher than the equilibrium value $\bar{X}_1$, the phase is oversaturated with $X$. However, comparing the values of the $\Phi (X_1)$ and $\Phi (\bar{X}_1)$ we see, that the deviation of the thermodynamic potential from its equilibrium value is of the higher order in $\bar{\varepsilon}$. On the other hand, if $\bar{\alpha} \neq 0$; $\bar{\eta} = 0$, see (3.29), similar to convective CHHCM, the final state after transition is slightly undersaturated by $X$ due to the presence of the field.

If both $\alpha$ and $\eta$ are non-zero, for the special value of $\bar{\alpha}$ (3.30), the velocity is zero, i.e. for the corresponding value of the applied field the transition front becomes static even for $X_2 \neq \frac{X_1}{2}$. Then the viscosity $\eta$ is self-consistently dropped out from the corrections to the stationary states, see (3.32).

Summing up, without viscosity and applied field the constant-velocity combined-transition-front model for CHHCM is not very instructive, both transitions should be too rigidly adjusted to each other (of course, for the non-constant-velocity-fronts situation could be quite different). However, the presence of the field and/or viscosity changes the situation; the concentration $X$ in the final state may deviate from its equilibrium value and even the transition may be stopped. On the other hand, for the CHS equation the influence of the non-equilibrium transition, i.e. of the reaction system, is much stronger. The transition front may be stopped, or even reversed both by changing the stationary states of the reaction system and by the field. The final state may be under-, or oversaturated, creating non-equilibrated phases. So, theoretically there is a possibility of engineering a material with some special properties, which differ from the properties of the equilibrium phase. Of course such a situation will last till the reservoir components will be substantially exhausted. Then the stationary state of the reaction system will be, naturally, destroyed. However, the destruction may be stopped, or at least slowed down, e.g., by lowering the temperature of the system.

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