Effective Macroscopic Interfacial Transport Equations in Strongly Heterogeneous Environments for General Homogeneous Free Energies

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Abstract

We study phase field equations in perforated domains for arbitrary free energies. These equations have found numerous applications in a wide spectrum of both science and engineering problems with homogeneous environments. Here, we focus on strongly heterogeneous materials with perforations such as porous media. To the best of our knowledge, we provide the first derivation of upscaled equations for general free energy densities. In view of the versatile applications of phase field equations, we expect that our study will lead to new modelling and computational perspectives for interfacial transport and phase transformations in strongly heterogeneous environments.

Keywords: Phase field models, multiscale analysis, homogenization, porous media, wetting

1. Introduction: Phase field formulation in heterogeneous media

Our starting point is the widely accepted diffuse-interface formulation \cite{1} describing the dynamics of interfaces between different phases. This formulation captures different thermodynamic states of a system by a continuous macroscopic variable obtained from averaged microscopic degrees of freedom. Such a macrovariable represents a locally conserved order parameter, denoted as $\phi$, which defines different phases as local equilibrium limiting values of a free energy associated with the system under consideration.

Diffuse interface formulations show a high versatility which is further extended due to increasing computational power. This leads continuously to new and increasingly complex scientific and engineering applications such as more realistic descriptions for the computation of transport in porous media \cite{2} which represents a high-dimensional multiscale problem with many numerical challenges \cite{3}. Our main result here is the systematic and general derivation of effective macroscopic equations which reliable account for multiple phases invading strongly heterogeneous environments such as porous materials.

The physical basis of the diffuse interface formulation relies on the following class of abstract energy densities

$$e(\phi) := \frac{1}{\lambda} F(\phi) + \frac{\lambda}{2} |\nabla \phi|^2.$$ \hfill (1)

The free energy density $F$ defines equilibrium phases $\phi_i, i = 1, 2, \ldots, M$ as $M \in \mathbb{N}$ local minima and the gradient term $\frac{\lambda}{2} |\nabla \phi|^2$ penalizes the interfacial area between these equilibrium phases. In thermodynamic contexts, $F$ represents the (Helmholtz) free energy density $F(\phi) := U - TS$, where $U$ is the internal energy, $T$ is the temperature, and $S$ is the entropy. Popular examples include the energy of regular solutions (also known as the Flory-Huggins energy \cite{4}). The regular solution theory plays a crucial role in many important applications such as ionic melts \cite{5}, water sorption in porous solids \cite{6}, and micellization in binary surfactant mixtures \cite{7}. Also wetting phenomena, often studied using classical sharp-interface approximations, e.g. \cite{8}, are also described by phase-field equations \cite{9,10,11} which have been extended to include electric fields (so-called electrowetting, e.g. \cite{12}). This energy-functional based framework has also been applied in image processing such as inpainting, see e.g. \cite{13}.

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In a previous study \cite{14}, we focused on a specific form of the homogeneous free energy density and we recently extended it towards Stokes flow \cite{15}. Here, we provide an upscaling for $H^{-1}$-gradient flows of arbitrary free energy densities based on a Taylor expansion of the free energy density at the effective upscaled solution. Before we can state our main result, we formulate the basic setting to study general interfacial dynamics.

(a) **Homogeneous domains $\Omega$.** The total (Ginzburg-Landau/Cahn-Hilliard) energy is defined by $E(\phi) := \int_{\Omega} e(\phi) \, dx$ with density $\underline{1}$ on a bounded domain $\Omega \subset \mathbb{R}^d$ with smooth boundary $\partial \Omega$ and $1 \leq d \leq 3$ denotes the spatial dimension. It is well accepted that thermodynamic equilibrium can be achieved by minimizing the energy $E$ supplemented by a possible wetting boundary contribution $\int_{\partial \Omega} g(x) \, dx$ for $g(x) \in H^{3/2}(\partial \Omega)$. Mass conservation for this minimization can be generally achieved with a $H^{-1}$-gradient flow associated to $E(\phi)$, i.e.,

\[
(\text{Homogeneous case}) \quad \frac{\partial}{\partial t} \phi = \text{div} \left( \tilde{M} \nabla \left( \frac{1}{\lambda} f(\phi) - \lambda \Delta \phi \right) \right) \quad \text{in} \; \Omega_T, \tag{2}
\]

together with the boundary conditions $\nabla_n \phi := n \cdot \nabla \phi = g(x)$ on $\partial \Omega_T$, and $\nabla_n \Delta \phi = 0$ on $\partial \Omega_T$, where $\Omega_T := \Omega \times [0, T[$, $\partial \Omega_T := \partial \Omega \times ]0, T[$, $\phi$ satisfies the initial condition $\phi(x, 0) = \psi(x)$, and $\tilde{M} = \{m_{ij}\}_{1 \leq i, j \leq d}$ denotes a mobility tensor with real and bounded elements $m_{ij} > 0$. This equation serves as a prototype for interfacial dynamics [e.g. \cite{16}] and phase transformation [e.g. \cite{1}] under homogeneous Neumann boundary conditions, i.e., $g = 0$, and free energy densities $F$.

(b) **Heterogeneous/perforated domains $\Omega^\varepsilon$.** Our main focus concentrates on \underline{1} in a perforated domain $\Omega^\varepsilon \subset \mathbb{R}^d$ instead of a homogeneous $\Omega \subset \mathbb{R}^d$. The parameter $\varepsilon = \frac{\ell}{T} > 0$ is called heterogeneity where $\ell$ represents the characteristic pore size and $L$ is the macroscopic length of the porous medium, see Figure \underline{1}. Here, we can define the porous medium by a reference pore/cell $Y := [0, \ell_1] \times [0, \ell_2] \times \cdots \times [0, \ell_d]$. For simplicity, we set $\ell_1 = \ell_2 = \cdots = \ell_d = 1$. The pore ($\Omega^\varepsilon$) and the solid phase ($B^\varepsilon$) are defined by

\[
\Omega^\varepsilon := \bigcup_{\mathbf{z} \in \mathbb{Z}^d} \varepsilon \{(Y^1 + \mathbf{z}) \cap \Omega \}, \quad B^\varepsilon := \bigcup_{\mathbf{z} \in \mathbb{Z}^d} \varepsilon \{(Y^2 + \mathbf{z}) \cap \Omega \} = \Omega \setminus \Omega^\varepsilon, \tag{3}
\]

where the subsets $Y^1, Y^2 \subset Y$ are such that $\Omega^\varepsilon$ is a connected set. The set $Y^1 \subset Y$ represents the pore phase (e.g. liquid or gas phase in wetting problems), see Figure \underline{1}. Here, we can rewrite (2) as the following microscopic porous media problem

\[
(\text{Micro-pore equation}) \quad \left\{ \begin{array}{ll}
\partial_t \phi_{\varepsilon} = \text{div} \left( \tilde{M} \nabla \left( -\lambda \Delta \phi_{\varepsilon} + \frac{1}{\varepsilon} f(\phi_{\varepsilon}) \right) \right) & \text{in} \; \Omega^\varepsilon_T, \end{array} \right. \tag{4}
\]

with the boundary ($\nabla_n \phi_{\varepsilon} := n \cdot \nabla \phi_{\varepsilon} = 0$ on $\partial \Omega^\varepsilon_T$, $\nabla_n \Delta \phi_{\varepsilon} = 0$ on $\partial \Omega^\varepsilon_T$) and initial ($\phi_{\varepsilon}(x, 0) = \psi(x)$ on $\Omega^\varepsilon$) conditions. Our main objective is the derivation of a systematic and reliable homogenized/upscaled phase field formulation valid for general energy densities \cite{11} by passing to the limit $\varepsilon \to 0$ in (4). We formally achieve this by asymptotic multiscale expansions \cite{17, 18}.

The main results are stated in Section \underline{2} and subsequently justified in Section \underline{3}.

Figure 1: **Left:** Strongly heterogeneous/perforated material as a periodic covering of reference cells $Y := [0, \ell]^d$. **Top,** middle: Definition of the reference cell $Y = Y^1 \cup Y^2$ with $\ell = 1$. **Right:** The "homogenization limit" $\varepsilon := \frac{\ell}{T} \to 0$ scales the perforated domain such that perforations become invisible on the macroscale.
2. Main results

Before we state our main result of effective macroscopic phase field equations (including the Cahn-Hilliard equation) which is valid for arbitrary energy densities \([1]\), we introduce the following scale separation property of the chemical potential.

**Definition 2.1.** (Scale separation) We say that the chemical potential is scale separated if and only if

\[
\frac{\partial \mu(\phi_0(x))}{\partial x_k} = \begin{cases} 
\begin{array}{ll}
0 & \text{on the reference cell } Y, \\
\frac{\partial \mu(\phi_0(x))}{\partial x_k} & \text{on the macroscopic } \Omega,
\end{array}
\end{cases}
\]

where \(\phi_0(x)\) is the upscaled/slow variable solving the upscaled phase field equation.

**Remark 2.2.** Definition 2.1 accounts for the problem specific separation between the large (macroscopic) scale \(x\) with slow processes and the small (microscopic) scale \(y\) with fast processes. \(\diamondsuit\)

In the homogenization/upscaling of nonlinear equations, Definition 2.1 appears naturally in the sense that it leads to the same class of equations on the macroscale as on the microscale and that it guarantees that resulting cell problems are well-posed [15, 19, 20]. These considerations together with the splitting strategy [14, 15], which decouples 4-th order problems [4] into two 2-nd order equations, allow us to state our

**Main Result:** (Upscaled Cahn-Hilliard equations) Suppose that \(\psi(x) \in H^2_F(\Omega)\). For scale separated chemical potentials \(\mu_0 = \nabla \phi E(\phi_0)\) (Definition 2.1), the microscopic porous media formulation [4] can be effectively and reliably approximated by the following macroscopic problem,

\[
\text{(Upscaled equation)} \quad \left\{ \begin{array}{l}
\theta_1 \frac{\partial \hat{\mu}}{\partial t} = \Delta \hat{M}_\phi / \lambda \nabla f(\phi_0) + \frac{1}{\eta_1} \Delta \left( M_\mu \nabla \left( \hat{D} \nabla \phi_0 \right) \right) \quad \text{in } \Omega_T, \\
\hat{M}_\phi = \left\{ m_{ik}^\phi \right\}_{1 \leq i, k \leq d} \text{ and } M_\mu = \left\{ m_{ik}^\mu(x) \right\}_{1 \leq i, k \leq d} \text{ are defined by}
\end{array} \right.
\]

\[
\left\{ \begin{array}{l}
d_{ik} \ := \ \frac{1}{|Y|} \sum_{j=1}^d \int_{Y_1} \left( \delta_{ik} - \delta_{ij} m_{ij}^\mu \right) dy, \\
m_{ik}^\phi \ := \ \frac{1}{|Y|} \sum_{j=1}^d \int_{Y_1} \left( m_{ik} - m_{ij} \frac{\partial \xi_k}{\partial y_j} \right) dy, \\
m_{ik}^\mu(x) \ := \ \frac{1}{|Y|} \sum_{j=1}^d \int_{Y_1} \left( m_{ik} - m_{ij} \frac{\partial \xi_k}{\partial y_j} \right) dy.
\end{array} \right.
\]

The corrector functions \(\xi_k^w \in H^1_{\text{per}}(Y^1)\) and \(\xi_k^\mu \in L^2(\Omega; H^1_{\text{per}}(Y^1))\) for \(1 \leq k \leq d\) solve in the distributional sense the following reference cell problems

\[
\left\{ \begin{array}{l}
\begin{cases}
- \sum_{i,j,k=1}^d \frac{1}{\partial y_i} 
\left( m_{ik} - m_{ij} \frac{\partial \xi_k^w}{\partial y_j} \right) 
= - \sum_{k,i,j=1}^d \frac{1}{\partial y_i} 
\left( m_{ik} - m_{ij} \frac{\partial \xi_k^w}{\partial y_j} \right) 
\end{cases} 
\text{in } Y^1, \\
\sum_{i,j,k=1}^d n_i \left( m_{ij} \frac{\partial \xi_k^w}{\partial y_j} - m_{ik} \right) + \left( m_{ik} - m_{ij} \frac{\partial \xi_k^w}{\partial y_j} \right) = 0 
\text{on } \partial Y^1 \cap \partial Y^2,
\end{array} \right.
\]

\(\xi_k^w(y)\) is \(Y\)-periodic and \(M_{Y^1}(\xi_k^w) = 0\).

\[
\left\{ \begin{array}{l}
\begin{cases}
- \sum_{i,j,k=1}^d \frac{1}{\partial y_i} 
\left( \delta_{ik} - \delta_{ij} \frac{\partial \xi_k^\mu}{\partial y_j} \right) 
= 0 
\end{cases} 
\text{in } Y^1, \\
\sum_{i,j=1}^d n_i \left( \delta_{ij} \frac{\partial \xi_k^\mu}{\partial y_j} - \delta_{ik} \right) = 0 
\text{on } \partial Y^1,
\end{array} \right.
\]

\(\xi_k^\mu(y)\) is \(Y\)-periodic and \(M_{Y^1}(\xi_k^\mu) = 0\).

The expression \(\nabla \phi E(\phi)\) denotes the Fréchet derivative of \(E\) with respect to \(\phi\). The upscaled equations show the mathematically and physically convincing feature that they preserve the structure from the microscopic formulation except for the effective correction tensors [6]. For a rigorous error quantification, we refer the interested reader to [21].

**Remark 2.3.** i) For an isotropic mobility, i.e., \(\hat{M} := m\hat{I}\) where \(\hat{I}\) is the identity matrix, we have \(\xi_k^w = \xi_k^\phi\). In this case, one can find computational results in Ref. [22] for the cell problems. \(\diamondsuit\)
3. Basic steps in the derivation using formal asymptotic expansions

We introduce the micro-scale $\xi: y \in Y$ and apply the standard multiscale property for spatial differentiation $\frac{\partial f(x)}{\partial x_i} = \frac{1}{\epsilon} \frac{\partial f}{\partial y_j}(x, x/\epsilon) + \frac{\partial f}{\partial x_i}(x, x/\epsilon)$, where $f(x) = f(x, y)$ is an arbitrary function depending on two variables $x \in \Omega$, $y \in Y$. The Laplace operators $\Delta$ and $\text{div}(\hat{M}\nabla)$ then can be written as follows,

$$
\begin{align*}
A_0 &= -\sum_{i,j=1}^{d} \frac{\partial}{\partial y_j} \left( \delta_{ij} \frac{\partial}{\partial y_i} \right), \\
A_1 &= -\sum_{i,j=1}^{d} \frac{\partial}{\partial x_i} \left( \delta_{ij} \frac{\partial}{\partial y_j} \right) + \frac{\partial}{\partial y_j} \left( \delta_{ij} \frac{\partial}{\partial x_i} \right), \\
A_2 &= -\sum_{i,j=1}^{d} \frac{\partial}{\partial x_j} \left( \delta_{ij} \frac{\partial}{\partial y_i} \right),
\end{align*}
$$

such that we can identify $A_\epsilon := \epsilon^{-2} A_0 + \epsilon^{-1} A_1 + A_2 = \Delta$ and correspondingly $B_\epsilon = \text{div}(\hat{M}\nabla)$. We account for the multiscale nature of strongly heterogeneous environments [19, 17, 18] by the following ansatz

$$
\xi^\epsilon \approx \xi_0(x, y, t) + \epsilon \xi_1(x, y, t) + \epsilon^2 \xi_2(x, y, t) + \ldots, \quad \text{for } \xi \in \{w, \phi\}.
$$

Before we can insert [9] into the microscopic formulation [4], we need to approximate the derivative of the nonlinear homogeneous free energy $f := F'$ by a Taylor expansion of the form

$$
f(\phi^\epsilon) \approx f(\phi_0) + f'(\phi_0)(\phi^\epsilon - \phi_0) + \frac{1}{2} f''(\phi_0)(\phi^\epsilon - \phi_0)^2 + O((\phi^\epsilon - \phi_0)^3),
$$

where $\phi_0$ denotes the leading-order term in [9]. Substituting [9] and [10] into [4], which we split into two second order problems as suggested in [14], and using [8] provides the following sequence of problems

$$
O(\epsilon^{-2}): \quad \begin{cases} 
B_0 [\lambda w_0 + 1/\lambda f(\phi_0)] = 0, & \text{in } Y^1, \\
A_0 \phi_0 = 0, & \text{under no flux b.c. and with } w_0 \text{ is } Y^1\text{-periodic}, \\
\nabla_n \phi_0 = 0 & \text{on } \partial Y_w^1 \cap \partial Y_w^2 \text{ and } \phi_0 \text{ } Y^1\text{-periodic},
\end{cases}
$$

$$
O(\epsilon^{-1}): \quad \begin{cases} 
B_0 [\lambda w_1 + 1/\lambda f'(\phi_0) \phi_1] = -B_1 [\lambda w_0 + 1/\lambda f(\phi_0)] & \text{in } Y^1, \\
A_0 \phi_1 = -A_1 \phi_0 & \text{under no flux b.c. and } w_1 \text{ is } Y^1\text{-periodic}, \\
\nabla_n \phi_1 = 0 & \text{on } \partial Y_w^1 \cap \partial Y_w^2 \text{ and } \phi_1 \text{ } Y^1\text{-periodic},
\end{cases}
$$

$$
O(\epsilon^0): \quad \begin{cases} 
B_0 [\lambda w_2 + \frac{1}{\chi} \left( \frac{1}{2} f''(\phi_0) \phi_1^2 + f'(\phi_0) \phi_2 \right)] = -B_2 [\lambda w_0 + 1/\lambda f(\phi_0)] + B_1 [\lambda w_1 - 1/\lambda f'(\phi_0) \phi_1] - \partial_t A_2^{-1} w_0 & \text{in } Y^1, \\
A_0 \phi_2 = -A_2 \phi_0 - A_1 \phi_1 + w_0 & \text{under no flux b.c. and with } w_2 \text{ is } Y^1\text{-periodic}, \\
\nabla_n \phi_2 = g_t \text{ on } \partial Y_w^1 \cap \partial Y_w^2 \text{ and } \phi_2 \text{ } Y^1\text{-periodic},
\end{cases}
$$

The first problem [11] immediately implies that the leading-orders $\phi_0$ and $w_0$ are independent of the microscale $y$ [19]. This suggests the following ansatz for $w_1$ and $\phi_1$, i.e.,

$$
w_1(x, y, t) = -\sum_{k=1}^{d} \xi_k^\epsilon(y) \frac{\partial w_0}{\partial x_k}(x, t), \quad \phi_1(x, y, t) = -\sum_{k=1}^{d} \xi_k^\epsilon(y) \frac{\partial \phi_0}{\partial x_k}(x, t) = \phi_1(x, y, t).
$$

Inserting [14] into [12] provides an equation for the correctors $\xi_k^\epsilon$ and $\xi_k^\phi$. The resulting equation for $\xi_k^\phi$ can be immediately written for $1 \leq k \leq d$ as,

$$
\xi_\phi: \quad \begin{cases} 
-\sum_{i,j=1}^{d} \frac{\partial}{\partial y_i} \left( \delta_{ik} - \delta_{ij} \frac{\partial \xi_k^\phi}{\partial y_j} \right) = -\text{div} \left( e_k - \nabla_y \xi_k^\phi \right) = 0 & \text{in } Y^1,
\end{cases}
$$

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$$

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$$
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\end{cases}
$$

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$$

Inserting [14] into [12] provides an equation for the correctors $\xi_k^\epsilon$ and $\xi_k^\phi$. The resulting equation for $\xi_k^\phi$ can be immediately written for $1 \leq k \leq d$ as,
for $\xi^k(y)$ $Y$-periodic with $M_{Y^1}(\xi^k_0) = 0$ and associated boundary condition $n \cdot \left( \nabla \xi^k_0 + e_k \right) = 0$ on $\partial Y^1_w \cap \partial Y^2_w$.

To study (12), we first rewrite $B_0 \left[ f'(\phi_0) \phi_1 \right]$ and $B_1 f(\phi_0)$ as follows

$$B_0 \left[ f'(\phi_0) \phi_1 \right] = - \sum_{k,i,j=1}^d \frac{\partial}{\partial y_i} \left( m_{ij} \frac{\partial \xi^k_0}{\partial y_j} \frac{\partial f(\phi_0)}{\partial x_k} \right), \quad B_1 f(\phi_0) = \sum_{i,j=1}^d \frac{\partial}{\partial y_i} \left( m_{ij} \frac{\partial f(\phi_0)}{\partial x_j} \right).$$

Doing the same for $w_1$ and $w_0$ and using (14) leads then to

$$- \lambda \sum_{k,i,j=1}^d \frac{\partial}{\partial y_i} \left( m_{ij} \left( \frac{\partial x_k}{\partial x_j} - \frac{\partial \xi^k_w}{\partial y_j} \right) \right) \frac{\partial w_0}{\partial x_k} = 1/\lambda \sum_{k,i,j=1}^d \frac{\partial}{\partial y_i} \left( m_{ij} \left( \frac{\partial x_k}{\partial x_j} - \frac{\partial \xi^k_0}{\partial y_j} \right) \right) \frac{\partial f(\phi_0)}{\partial x_k},$$

in $Y^1$. Since the chemical potential $\mu(\phi) = \frac{\delta E(\phi)}{\delta \phi}$ is scale separated, we finally obtain the reference cell problem for $\xi^k_{w}$, $1 \leq k \leq d$ for given $\xi^k_{w}$

$$\left\{ \begin{array}{l}
- \sum_{i,j,k=1}^d \delta_{ik} \frac{\partial}{\partial y_i} \left( m_{ik} - m_{ij} \frac{\partial \xi^k_0}{\partial y_j} \right) \\
= - \sum_{i,j,k=1}^d \frac{\partial}{\partial y_i} \left( m_{ik} - m_{ij} \frac{\partial \xi^k_w}{\partial y_j} \right) \end{array} \right. \text{ in } Y^1,$n

where $\xi^k_{w}(y)$ is $Y$-periodic with boundary condition $\sum_{i,j,k=1}^d n_i \left( \left( m_{ij} \frac{\partial \xi^k_w}{\partial y_j} - m_{ik} \right) + \left( m_{ik} - m_{ij} \frac{\partial \xi^k_w}{\partial y_j} \right) \right) = 0$ on $\partial Y^1_w \cap \partial Y^2_w$ and $M_{Y^1}(\xi^k_w) = 0$.

The last problem (13) then gives the upscaled equations by a solvability constraint, i.e., the Fredholm alternative. Hence, the solvability of (13) is achieved by setting

$$- \sum_{i,k=1}^d \int_{Y^1} \left[ \delta_{ik} \frac{\partial}{\partial x_i} \left( m_{ik} - \frac{\partial \xi^k_w}{\partial y_j} \right) \right] \frac{\partial^2 \phi_0}{\partial x_i \partial x_k} = \left| Y^1 \right| w_0 + \tilde{g}_0.$$

where $\tilde{g}_0 := - \frac{\partial}{\partial x_i} \int_{\partial Y^1} \left( a_1 \chi_{\partial Y^1_{w1}} + a_1 \chi_{\partial Y^1_{w2}} \right) \, dy \chi$. (19) can be written compactly as $- \Delta_D \phi_0 := - \text{div} \left( \hat{D} \nabla \phi_0 \right) = \theta_1 w_0 + \tilde{g}_0$ by (6). Applying the same ideas to equation (13) leads to

$$\int_{Y^1} \left\{ - \lambda (B_2 w_0 + B_1 w_1) - \frac{1}{\lambda} B_1 \left[ f'(\phi_0) \phi_1 \right] - \frac{1}{\lambda} B_2 f(\phi_0) - \partial_t A^{-1}_2 w_0 \right\} \, dy = 0,$$

where the first two terms can be rewritten with (6) by $\int_{Y^1} (B_2 w_0 + B_1 w_1) \, dy = \text{div} \left( \hat{M}_w \nabla w_0 \right)$. The third integrand in (20) becomes $-B_1 \left[ f'(\phi_0) \phi_1 \right] = - \sum_{i,j=1}^d m_{ij} \sum_{k=1}^d \frac{\partial \xi^k_0}{\partial y_j} \frac{\partial^2 f(\phi_0)}{\partial x_i \partial x_k}$, where we applied the chain rule $\frac{\partial^2 f(\phi_0)}{\partial x_i \partial x_j} = f''(\phi_0) \frac{\partial f(\phi_0)}{\partial x_i} \frac{\partial f(\phi_0)}{\partial x_j} + f'(\phi_0) \frac{\partial^2 f(\phi_0)}{\partial x_i \partial x_j}$ and integration by parts. Add now the term $-B_2 f(\phi_0)$ and using (6) finally gives $-B_1 \left[ f'(\phi_0) \phi_1 \right] - B_2 f(\phi_0) = \text{div} \left( \hat{M}_w \nabla f(\phi_0) \right)$.

These considerations finally lead to the following effective equation for $\phi_0$, i.e.,

$$\theta_1 \frac{\partial \delta \phi_0}{\partial t} = \text{div} \left( \hat{M}_w / \lambda \nabla f(\phi_0) \right) + \frac{\lambda}{\theta_1} \text{div} \left( \hat{M}_w \nabla \left( \text{div} \left( \hat{D} \nabla \phi_0 \right) - \tilde{g}_0 \right) \right).$$

In the case where the homogeneous free energy $F$ is the classical double-well potential $F(\phi) = 1/4(\phi^2 - 1)^2$, one can verify the solvability of (21) along with the arguments in (23).

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