SHARP-INTERFACE APPROACH FOR SIMULATING SOLID-STATE DEWETTING IN THREE DIMENSIONS

WEI JIANG∗, QUAN ZHAO†, AND WEIZHU BAO‡

Abstract. The problem of simulating solid-state dewetting of thin films in three dimensions (3D) by using a sharp-interface approach is considered in this paper. Based on the thermodynamic variation, a speed method is used for calculating the first variation to the total surface energy functional. The speed method shares more advantages than the traditional use of parameterized curves (or surfaces), e.g., it is more intrinsic and its variational structure (related with Cahn-Hoffman \( \xi \)-vector) is clearer and more direct. By making use of the first variation, necessary conditions for the equilibrium shape of the solid-state dewetting problem is given, and a kinetic sharp-interface model which includes the surface energy anisotropy is also proposed. This sharp-interface model describes the interface evolution in 3D which occurs through surface diffusion and contact line migration. By solving the proposed model, we perform lots of numerical simulations to investigate the evolution of patterned films, e.g., the evolution of a short cuboid and pinch-off of a long cuboid. Numerical simulations in 3D demonstrate the accuracy and efficacy of the sharp-interface approach to capture many of the complexities observed in solid-state dewetting experiments.

Key words. Solid-state dewetting, surface diffusion, Cahn-Hoffman \( \xi \)-vector, shape derivative, thermodynamic variation.

AMS subject classifications. 74G65, 74G15, 74H15, 49Q10

1. Introduction. Driven by capillarity effects, solid thin films sitting on a substrate are rarely stable and could exhibit complex morphological changes, e.g., faceting [28, 29, 60], edge retraction [58, 13, 61, 62, 32], pinch-off [23, 31], fingering instabilities [30, 10, 18] and so on. This phenomenon, known as solid-state dewetting [27], has been widely observed in many thin film/substrate systems [28, 29, 32, 48]. From one side, solid-state dewetting can be deleterious by fabricating the thin film structures, e.g., microelectronic and optoelectronic devices, thus destroying the reliability of the devices; on the other side, it is advantageous and can be positively used to produce the well-controlled formation of an array of micro-/nanoscale particles, e.g., used in sensors [37, 2] and as catalysts for carbon [44] and semiconductor nanowire growth [66]. Recently, solid-state dewetting has attracted considerable interest, and has been widely studied by many experimental (e.g., [1, 20, 40, 39, 34, 43, 61]) and theoretical (e.g., [1, 5, 17, 22, 27, 51, 53, 18, 55, 30, 60]) research groups. The understanding of its equilibrium patterns and kinetic morphology evolution characteristics could provide important knowledge to develop new experimental methods in order to control solid-state dewetting [35], and enhance its potential applications in thin film technologies.

Modeling solid-state dewetting has been one of active research areas and become increasingly urgent in decades. In general, surface diffusion and contact line mi-

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migration have been recognized as the two main kinetic features for the evolution of solid-state dewetting \[4, 26\]. In 1986, Srolovitz and Safran \[51\] proposed a simplified sharp-interface model to study the hole growth during the dewetting under the three assumptions, i.e., isotropic surface energy, small slope profile and cylindrical symmetry. Based on the above model, Wong et al. designed a “marker particle” numerical scheme to investigate the two-dimensional retraction of a discontinuous film (a film with a step) and the evolution of a perturbed cylindrical wire on a substrate \[58, 14\]. These earlier studies were focused on the isotropic surface energy, although recent experiments have demonstrated that the crystalline anisotropy could play important roles in solid-state dewetting. To include the surface energy anisotropy, many approaches have been proposed in recent years, such as a discrete model \[13\], a kinetic Monte Carlo model \[42, 15\], a crystalline model \[9, 65\] and continuum models based on partial differential equations \[3, 25, 20, 55\]. From a mathematical perspective, theoretical solid-state dewetting studies can be categorized into two major problems: one focuses on the equilibrium of solid particles on substrates \[4, 33\]; the other focuses on investigating the kinetic evolution of solid-state dewetting \[25, 26, 55\]. In this paper, we will develop a sharp-interface approach for studying these problems about solid-state dewetting in 3D.

Under isothermal conditions, the equilibrium shape for a free-standing solid particle can be formulated by minimizing the interfacial energy subject to the constraint of a constant volume:

\[
\min_{\Omega} W := W(S) = \int_S \gamma(n) \, dS \quad \text{s.t.} \quad |\Omega| = \text{const},
\]

where \(\Omega \subseteq \mathbb{R}^3\) is the enclosed domain by a closed surface \(S\), and \(\gamma(n)\) is the surface energy (density) with \(n = (n_1, n_2, n_3)^T\) representing the crystallographic orientation. Based on the \(\gamma\)-plot, the equilibrium shape can be geometrically constructed via the well-known Wulff (Gibbs-Wulff) construction \[59\]. The resulted Wulff shape, is the inner convex region bounded by all planes that are perpendicular to orientation \(n\) and at a distance of \(\gamma(n)\) from the origin. The Winterbottom construction \[57, 5\] was subsequently proposed to handle with the case about particles on substrates by truncating the Wulff shape with a flat plane, and where the Wulff shape is truncated depends on the wettability of the substrate. Meanwhile, many theories \[7, 8\] demonstrated that the derivative of \(\gamma(n)\) plays an important role in investigating equilibrium and kinetic problems for solid particles with anisotropic surface energies. In 1972, Cahn and Hoffman developed the theory of \(\xi\)-vector \[22, 7\] to describe the surface energy anisotropy of solid materials. It is defined based on a homogeneous extension of \(\gamma(n)\):

\[
\xi(n) = \nabla \tilde{\gamma}(p) \bigg|_{p=n}, \quad \text{with} \quad \tilde{\gamma}(p) = |p| \gamma\left(\frac{p}{|p|}\right), \quad \forall p \in \mathbb{R}^3 \setminus \{0\},
\]

where \(|p| := \sqrt{p_1^2 + p_2^2 + p_3^2}\) for \(p = (p_1, p_2, p_3)^T \in \mathbb{R}^3\). Under this extension, \(\tilde{\gamma}(p)\) satisfies

\[
\tilde{\gamma}(\lambda p) = |\lambda| \tilde{\gamma}(p), \quad \nabla \tilde{\gamma}(p) \cdot p = \tilde{\gamma}(p), \quad \forall \lambda \neq 0, p \in \mathbb{R}^3 \setminus \{0\}.
\]

Compared to the traditional use of scalar function \(\gamma\) (or \(\gamma\)-plot), \(\xi\)-vector formulation has some advantages in the description of equilibrium shapes and thermodynamic evolution for crystalline interfaces \[26, 56\]. From \(\xi \cdot n = \gamma(n)\), we have
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Fig. 1.1. \(\gamma\)-plot, \(1/\gamma\)-plot and \(\xi\)-plot for different surface energy anisotropies: (a) isotropic surface energy; (b) cubic anisotropic surface energy defined as \(\gamma(n) = 1 + 0.3(n_1^4 + n_2^4 + n_3^4)\); (c) ellipsoidal surface energy \(\gamma(n) = \sqrt{4n_1^2 + n_2^2 + n_3^2}\); (d) “cusped” surface energy defined as \(\gamma(n) = |n_1| + |n_2| + |n_3|\).

magnitude of the normal component for \(\xi\) equals to \(\gamma(n)\). Meanwhile, \(\xi\)-plot shares similar geometry with the Wulff shape, and it can be regarded as a mathematical representation of the equilibrium shape when its \(1/\gamma\)-plot is convex (i.e., weakly anisotropic). Fig. 1.1 depicts the \(\gamma\)-plot, \(1/\gamma\)-plot and \(\xi\)-plot for four different types of surface energy anisotropies: (a) isotropic surface energy, i.e., \(\gamma(n) \equiv 1\); (b) cubic surface energy \(\gamma(n) = 1 + a(n_1^4 + n_2^4 + n_3^4)\) with \(a\) representing the degree of anisotropy; (c) ellipsoidal surface energy \(\gamma(n) = \sqrt{a_1^2n_1^2 + a_2^2n_2^2 + a_3^2n_3^2}\); (d) “cusped” surface energy defined as \(\gamma(n) = |n_1| + |n_2| + |n_3|\). In the application of materials science, the surface energy could be piecewise smooth and have some “cusped” points, where it is not differentiable. A typical example is the “cusped” surface energy defined above. For these cases, we can regularize the surface energy with a small parameter \(0 < \varepsilon \ll 1\) to ensure the usage of sharp-interface approach proposed in this paper, i.e.,

\[
\gamma(n) = \sqrt{\varepsilon^2 + (1 - \varepsilon^2)n_1^2} + \sqrt{\varepsilon^2 + (1 - \varepsilon^2)n_2^2} + \sqrt{\varepsilon^2 + (1 - \varepsilon^2)n_3^2}.
\]

The Cahn-Hoffman \(\xi\)-vector has been recently utilized to describe the solid-state dewetting problem in two dimensions (2D) \cite{26}. Based on the thermodynamic variation, the authors derived a sharp-interface approach via the \(\xi\)-vector formulation for describing the kinetic evolution of solid-state dewetting in 2D. In this approach, the moving interface is described as a parametrization over a time-independent domain, and the variation is performed by considering an infinitesimal perturbation with respect to an open interface curve coupled with contact points \cite{26}. However, when we want to generalize this approach to 3D, we realize that it would be very different for calculating the thermodynamic variation for the 3D problem by using the approach
of parameterized surfaces. First, the calculations of the variation in 3D via surface parametrization approach would become complicated, extremely tedious and a nightmare, and it unavoidably involves in a lot of knowledge about differential geometry; Second, for the solid-state dewetting problem, the infinitesimal perturbation to a surface in the tangential direction plays an important role in investigating the contact line migration along the substrate \cite{25,4}, and it would make the calculations become more complicated; Third, complicated calculations often make people easily forget the nature of the problem, and we need to investigate and make use of the variational structure of the problem. These difficulties motivate us to look for a new approach to calculating the thermodynamic variation of solid-state dewetting in 3D. In the literature, the shape optimization problem is popular in the design of industrial structures. The speed method and shape derivatives have been widely utilized to perform the shape sensitivity analysis of shape optimization problems \cite{49,21,12}. This approach avoids the parametrization of a surface and is able to deal with perturbations along arbitrary directions, and it is the desired tool we are searching for.

Therefore, based on the $\xi$-vector formulation and the speed method, the objectives of this paper are as follows: (i) to calculate the thermodynamic variation of the energy functional for solid-state dewetting in 3D; (ii) to provide a rigorous derivation of the thermodynamic description of the equilibrium shape for solid-state dewetting in 3D; (iii) to develop a sharp-interface model which includes surface diffusion and contact line migration for simulating kinetic evolution of solid-state dewetting in 3D; and (iv) to present numerical simulations to investigate important characteristics of the morphological evolution for solid-state dewetting observed in experiments.

The rest of the paper is organized as follows. In Section 2, we briefly introduce the speed method and sharp derivatives, and then apply them for calculating the first variation of the total free energy functional. In Section 3, we rigorously derive the necessary conditions for the equilibrium shape and explicitly give an expression for the equilibrium shape by using a parametric formula. In Section 4, based on thermodynamic variation, a sharp-interface model is proposed for simulating solid-state dewetting of thin films in 3D. Subsequently, we perform some numerical simulations to demonstrate the accuracy and efficacy of our proposed model in Section 5. Finally, we draw some conclusions in Section 6.

2. Thermodynamic variation. The solid-state dewetting problem can be illustrated as Fig. 2.1, where a solid thin film (in blue) can dewet or agglomerate on a flat rigid substrate (in gray) due to capillarity effects. The total interfacial free energy of the system can be written as \cite{11,26}

\[
W = \int_{S_{FV}} \gamma_{FV} \, dS_{FV} + \int_{S_{FS}} \gamma_{FS} \, dS_{FS} + \int_{S_{VS}} \gamma_{VS} \, dS_{VS},
\]

where $S_{FV} := S$, $S_{FS}$ and $S_{VS}$ represent the film/vapor, film/substrate and vapor/substrate interfaces, respectively, and $\gamma_{FV}$, $\gamma_{FS}$ and $\gamma_{VS}$ represent the corresponding surface energy densities. In solid-state dewetting problems, we often assume that $\gamma_{FS}$, $\gamma_{VS}$ are two constants, and $\gamma_{FV}$ is a function of the orientation of the film/vapor interface, i.e., $\gamma_{FV} := \gamma(n)$ with $n$ representing the unit normal vector of the film/vapor interface, which points outwards to the vapor phase. The film/vapor interface is here described by an open two-dimensional surface $S$ with boundary $\Gamma$ (i.e., the contact line), which is a closed plane curve on the flat substrate $S_{sub}$. 


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Substrate
Film
Vapor
\( \gamma \)
\( \gamma_{FS} \)
\( \gamma_{VS} \)

\( \gamma_{RV} = \gamma(\mathbf{n}) \)

\( \mathbf{n} \)
\( \mathbf{c}_T \)
\( \mathbf{\tau}_T \)

\( \text{Fig. 2.1. A schematic illustration of the solid-state dewetting of a solid thin film (in blue) on a flat, rigid substrate (in gray) in 3D.} \)

Assume that we consider a bounded domain with size \( L_x \times L_y \) on the substrate (shown in Fig. 2.1), and we label the surface area enclosed by the contact line \( \Gamma \) as \( A(\Gamma) \), then the total interfacial free energy of the system can be calculated as

\[
W = \int_{S_{FS}} \gamma_{FS} \, dS_{FS} + \int_{S_{VS}} \gamma_{VS} \, dS_{VS} + \int_{S_{SV}} \gamma_{SV} \, dS_{SV}
\]

\[
= \int_S \gamma(\mathbf{n}) \, dS + A(\Gamma) \gamma_{FS} + (L_x L_y - A(\Gamma)) \gamma_{VS}
\]

\[
= \int_S \gamma(\mathbf{n}) \, dS + (\gamma_{FS} - \gamma_{VS}) A(\Gamma) + L_x L_y \gamma_{VS}.
\]

By dropping off the constant term \( L_x L_y \gamma_{VS} \), we can simplify the total interfacial free energy (still labeled as \( W \)) as the following two parts, i.e., the film/vapor interface energy term \( W_{\text{int}} \) and the substrate energy term \( W_{\text{sub}} \),

\[
W = W_{\text{int}} + W_{\text{sub}} = \int_S \gamma(\mathbf{n}) \, dS + (\gamma_{FS} - \gamma_{VS}) A(\Gamma).
\]

As shown in Fig. 2.1 we introduce three unit vectors \( \mathbf{n}_T \), \( \mathbf{\tau}_T \) and \( \mathbf{c}_T \), which are defined along the boundary \( \Gamma \). More precisely, \( \mathbf{n}_T \) is the outer unit normal vector of the plane curve \( \Gamma \) on the substrate \( S_{\text{sub}} \); \( \mathbf{\tau}_T \) is the unit tangent vector of \( \Gamma \) on the substrate plane \( S_{\text{sub}} \), which points anticlockwise when looking from top to bottom; \( \mathbf{c}_T \) is called as the co-normal vector, which is normal to \( \Gamma \) and tangent to the surface \( S \), and points downwards. For any point \( \mathbf{x} \in S \) (with \( \mathbf{x} = (x_1, x_2, x_3)^T \) or \( (x, y, z)^T \)), if we label \( T_{\mathbf{x}} S \) and \( N_{\mathbf{x}} S \) as the tangent and normal vector spaces to \( S \) at \( \mathbf{x} \), respectively, then the following properties are valid,

\[
\mathbf{\tau}_T(\mathbf{x}) \in T_{\mathbf{x}} S, \quad \mathbf{\tau}_T(\mathbf{x}) \parallel S_{\text{sub}}, \quad \mathbf{\tau}_T(\mathbf{x}) \perp \mathbf{c}_T(\mathbf{x}), \quad \forall \mathbf{x} \in \Gamma,
\]

\[
\mathbf{c}_T(\mathbf{x}) \in T_{\mathbf{x}} S, \quad \mathbf{n}_T(\mathbf{x}) \parallel S_{\text{sub}}, \quad \mathbf{n}_T(\mathbf{x}) \perp \mathbf{\tau}_T(\mathbf{x}), \quad \forall \mathbf{x} \in \Gamma.
\]

2.1. Differential operators on manifolds. To obtain the thermodynamic (or first) variation of the above shape functional \( W \), we start by introducing some basic
knowledge about surface calculus and differential geometry. For more details, readers can refer to [10] [11].

**Definition 2.1.** Suppose that $S \subset \mathbb{R}^3$ is a two-dimensional smooth manifold, and a function $f$ is defined on $S$ such that $f \in C^2(S)$. Let $\mathbf{n} = (n_1, n_2, n_3)^T$ be the unit outer normal vector of $S$, and $\bar{f}$ be an extension of $f$ in the neighbourhood of $S$ such that $f$ is differentiable, then the surface gradient of $f$ on $S$ is defined as

$$\nabla_S f = \nabla \bar{f} - (\nabla \bar{f} \cdot \mathbf{n}) \mathbf{n},$$

with $\nabla$ denoting the usual gradient in $\mathbb{R}^3$. It is easy to show that $\nabla_S f$ is independent of the extension of $f$ and only dependent on the value of $f$ on $S$. If we denote $\nabla_S$ as a vector operator

$$\nabla_S = (D_1, D_2, D_3)^T,$$

then we can easily obtain

$$D_i x_j = \delta_{ij} - n_i n_j, \quad \forall 1 \leq i, j \leq 3,$$

where $\mathbf{x} = (x_1, x_2, x_3)$ is the position vector of the surface and $\delta_{ij}$ is the Kronecker delta. The surface divergence of a vector-valued function $\mathbf{g} = (g_1, g_2, g_3)^T \in [C^1(S)]^3$ is defined as

$$\nabla_S \cdot \mathbf{g} = \sum_{i=1}^3 D_i g_i.$$

Moreover, the Laplace-Beltrami operator on $S$ can be expressed as

$$\Delta_S f = \nabla_S \cdot (\nabla_S f) = \sum_{i=1}^3 D_i D_i f.$$

In the definition of surface gradient, since the normal component has been subtracted from $\nabla \bar{f}$, $\nabla_S f$ can be viewed as the tangential component of $\nabla \bar{f}$, and thus we have $\nabla_S f \cdot \mathbf{n} = 0$ and $\nabla_S f(\mathbf{x}) \in T_x S$, $\forall \mathbf{x} \in S$. Note that it can be rigorously proved that Definition [2.1] is consistent with the conventional definition in differential geometry [10], and it generalizes the definition domain of surface divergence from the vector in tangent vector spaces to any vector in $\mathbb{R}^3$.

On the other hand, the integration by parts on an open smooth surface $S$ with smooth boundary $\Gamma$ reads as (see Theorem 2.10 in [10], and we omit the proof here)

$$\int_S \nabla_S f \, dS = \int_S f \, \mathcal{H} \, \mathbf{n} \, dS + \int_{\Gamma} f \, \mathbf{c}_\Gamma \, d\Gamma,$$

where $\mathbf{n}$ and $\mathbf{c}_\Gamma$ are the normal and co-normal vectors (shown in Fig. 2.1), respectively, and $\mathcal{H}$ is the mean curvature, which is defined as the surface divergence of the unit normal vector, i.e., $\mathcal{H} = \nabla_S \cdot \mathbf{n}$. Similarly, by the above equation and Definition [2.1] we can obtain the integration by parts about a vector field $\mathbf{F} = (f_1, f_2, f_3)^T \in \mathbb{R}^3$ on an open smooth surface $S$ with smooth boundary $\Gamma$,

$$\int_S \nabla_S \cdot \mathbf{F} \, dS = \int_S \mathcal{H} \mathbf{F} \cdot \mathbf{n} \, dS + \int_{\Gamma} \mathbf{F} \cdot \mathbf{c}_\Gamma \, d\Gamma.$$
If $F$ lies in the tangent vector space of $S$, i.e., $F \cdot n = 0$, then the second term vanishes. Furthermore, by using the product rule that $\nabla_s (fg) = g \nabla_s f + f \nabla_s g$, we can obtain

\begin{equation}
\int_S g \nabla_s f \, dS = -\int_S f \nabla_s g \, dS + \int_S f g \, dS + \int_{\Gamma} f g \, d\Gamma.
\end{equation}

In a simple case, if $S$ is a flat surface (i.e., $\mathcal{H} = 0$) with a plane boundary curve $\Gamma$, then Eq. (2.10) reduces to

\begin{equation}
\int_S \nabla_s f \, dS = \int_{\Gamma} f \, d\Gamma,
\end{equation}

which is the Gauss-Green theorem in the multivariable calculus, because $\nabla_s f$ collapses to the gradient of $f$ in 2D, and $c_r$ collapses to the unit outer normal vector of $\Gamma$.

### 2.2. The speed method and shape derivative.

In this section, the objective is to calculate the first variation of the energy (or shape) functional defined in (2.3). To this end, we first introduce an independent parameter $\epsilon \in [0, \epsilon_0]$ to parameterize a family of perturbations of a given domain $D \subset \mathbb{R}^3$, where the parameter $\epsilon$ controls the amplitude of the perturbation and $\epsilon_0$ is the maximum perturbation amplitude. Furthermore, we assume that the domain $D$ is of class $C^k$ with $k \geq 2$.

More precisely, we consider a domain $D \subset \mathbb{R}^3$ with a piecewise smooth boundary $\partial D$, then we can construct a family of transformations $T_{\epsilon}$, which are one-to-one, and $T_{\epsilon}$ maps from $\bar{D}$ onto $D$, i.e.,

\begin{equation}
T_{\epsilon} : \bar{D} \to D, \quad \epsilon \in [0, \epsilon_0),
\end{equation}

where $\epsilon$ is the small perturbation parameter. Generally, we assume that: (i) $T_{\epsilon}$ and $T_{\epsilon}^{-1}$ belong to $C^k(\bar{D}, \mathbb{R}^3)$ for all $\epsilon \in [0, \epsilon_0)$ with $k \geq 2$; and (ii) the mappings $\epsilon \to T_{\epsilon}(x)$ and $\epsilon \to T_{\epsilon}^{-1}(x)$ belongs to $C^1[0, \epsilon_0)$ for all $x \in \mathbb{R}^3$ (with $x = (x_1, x_2, x_3)^T$).

Given any point $X \in \bar{D}$ (with $X = (X_1, X_2, X_3)^T$) and $\epsilon \in [0, \epsilon_0)$, we can define the point $x = T_{\epsilon}(X)$ which moves along the trajectory. Here, the point $X$ represents the Lagrangian (or material) coordinate, while $x$ is the Eulerian (or actual) coordinate. Therefore, the speed vector field $V(x, \epsilon)$ at point $x$ is defined as

\begin{equation}
V(x, \epsilon) = \frac{\partial x}{\partial \epsilon}(T_{\epsilon}^{-1}(x), \epsilon).
\end{equation}

On the other hand, the transformation $T_{\epsilon}$ can be uniquely determined by the speed vector field $V$ via the following ordinary differential equation (ODE)

\begin{equation}
\begin{cases}
\frac{dx}{d\epsilon}(x, \epsilon) = V(x(X, \epsilon), \epsilon), \\
x(X(0), \epsilon) = X.
\end{cases}
\end{equation}

Therefore, the transformation $T_{\epsilon}$ and the smooth vector field $V$ are uniquely determined by each other. For a smooth vector field $V$, e.g., $V \in C(C^k(\bar{D}, \mathbb{R}^3); [0, \epsilon_0])$, the equivalence between the transformation $T_{\epsilon}$ and the speed vector field $V$ has been strictly established by Theorem 2.16 in [49]. In the following, we use $T_{\epsilon}(V)$ to denote the transformation associated with vector field $V$. For simplicity, we also denote $V_0 = V(X, 0)$.

Let $J(G)$ be a shape functional defined on a shape $G \subset \bar{D}$, where $G$ could be a three-dimensional domain (e.g., $\Omega$) or a two-dimensional manifold (e.g., a surface $S$).
The first variation of the functional $J(G)$ at $G$ in the direction of a speed vector field $V \in C(C^k(\bar{D}, D); [0, \epsilon_0))$ is given as the Eulerian derivative:

\begin{equation}
\delta J(G; V) = \lim_{\epsilon \to 0} \frac{J(G_{\epsilon}) - J(G)}{\epsilon},
\end{equation}

where $G_{\epsilon} = T_\epsilon(V)(G)$. To obtain the first variation and based on the transformation, we first define the material derivative and shape derivative of a function on a domain $\Omega$ or a surface $S$. For more details about the shape differential calculus, we refer to the book by Sokolowski and Zolesio \[49\].

**Definition 2.2.** (Material derivatives, Def. 2.71 and Def. 2.74 in \[49\]) The material derivative $\dot{\psi}(\Omega; V)$ of $\psi$ on a domain $\Omega$ in the direction of a speed vector field $V$ is defined as

\[ \dot{\psi}(\Omega; V) = \lim_{\epsilon \to 0} \frac{\psi(\Omega_{\epsilon}) \circ T_\epsilon(V) - \psi(\Omega)}{\epsilon}, \]

where for $X \in \Omega$, $\psi(\Omega_{\epsilon}) \circ T_\epsilon(V) = \psi(T_\epsilon(X))$.

Similarly, the material derivative $\dot{\varphi}(S; V)$ of $\varphi$ on a surface $S$ in the direction $V$ is defined as

\[ \dot{\varphi}(S; V) = \lim_{\epsilon \to 0} \frac{\varphi(S_{\epsilon}) \circ T_\epsilon(V) - \varphi(S)}{\epsilon}, \]

where for $X \in S$, $\varphi(S_{\epsilon}) \circ T_\epsilon(V) = \varphi(T_\epsilon(X))$.

**Definition 2.3.** (Shape derivatives, Def. 2.85 and Def. 2.88 in \[49\]) The shape derivative $\psi'(\Omega; V)$ of $\psi$ defined on a domain $\Omega$ in the direction $V$ is defined as

\[ \psi'(\Omega; V) = \dot{\psi}(\Omega; V) - \nabla \psi(\Omega) \cdot V_0. \]

Similarly, the shape derivative $\varphi'(S; V)$ of $\varphi$ defined on a surface $S$ in the direction $V$ is defined as

\[ \varphi'(S; V) = \dot{\varphi}(S; V) - \nabla_s \varphi(S) \cdot V_0. \]

**Proposition 2.1.** Let $\Omega \subset \mathbb{R}^d$ be a smooth bounded domain in $D$ with smooth boundary $\partial \Omega$, and $V$ be a speed vector field such that $V \in C(C^k(\bar{D}, D); [0, \epsilon_0))$. Suppose that $\psi = \psi(\Omega)$ is given such that the material derivative $\dot{\psi}(\Omega; V)$ and the shape derivative $\psi'(\Omega; V)$ exist. Then, the shape functional $J(\Omega) = \int_\Omega \psi(\Omega) d\Omega$ is shape differentiable and we have

\begin{equation}
\delta J(\Omega; V) = \int_\Omega \psi'(\Omega; V) d\Omega + \int_{\partial \Omega} \psi(\Omega) V_0 \cdot n d\Omega.
\end{equation}

**Proof.** By using the change of variables $x = T_\epsilon(V)(X)$ for $J(\Omega_{\epsilon})$, we have

\begin{equation}
J(\Omega_{\epsilon}) = \int_\Omega \psi(\Omega_{\epsilon}) \circ T_\epsilon(V) \zeta(x, \epsilon) d\Omega,
\end{equation}
where $\zeta(X, \epsilon) = \det(DT_\epsilon)$ with $DT_\epsilon = \left(\frac{\partial T_{ij}(V)(X)}{\partial X_k}(X)\right)_{i,j=1,2,3}$. By noting the fact that $\zeta(X, 0) = 1$, \(\frac{\partial \zeta(X, \epsilon)}{\partial \epsilon}\bigg|_{\epsilon=0} = \nabla \cdot \mathbf{V}_0\) (see Proposition 2.44 in [49]), we have

$$
\delta J(\Omega; \mathbf{V}) = \lim_{\epsilon \to 0} \frac{J(\Omega_\epsilon) - J(\Omega)}{\epsilon}
= \int_{\Omega} \left[\psi(\Omega; \mathbf{V})\zeta(X, \epsilon) + \psi(\Omega) \frac{\partial \zeta(X, \epsilon)}{\partial \epsilon}\bigg|_{\epsilon=0}\right] d\Omega
$$

(2.24)

$$
= \int_{\Omega} \psi(\Omega; \mathbf{V}) d\Omega + \int_{\partial \Omega} \psi(\Omega) \nabla \cdot \mathbf{V}_0 d\Omega.
$$

By using the definition of the shape derivative $\psi'(\Omega; \mathbf{V})$ of $\psi(\Omega)$ on a domain $\Omega$, i.e., $\psi'(\Omega; \mathbf{V}) = \dot{\psi}(\Omega; \mathbf{V}) - \nabla \psi(\Omega) \cdot \mathbf{V}_0$, and integration by parts for the second term of Eq. (2.24), we immediately obtain

$$
\delta J(\Omega; \mathbf{V}) = \int_{\Omega} \psi'(\Omega; \mathbf{V}) d\Omega + \int_{\partial \Omega} \psi(\Omega) \mathbf{V}_0 \cdot \mathbf{n} d\Omega,
$$

which completes the proof. \(\square\)

**Remark 2.1.** If $\mathbf{V}_0 \cdot \mathbf{n} = 0$ on the boundary $\partial \Omega$, we obtain that the first variation of the functional reduces to

$$
\delta J(\Omega; \mathbf{V}) = \int_{\Omega} \psi'(\Omega; \mathbf{V}) d\Omega.
$$

**Remark 2.2.** The material derivative can be regarded as the derivative with respect to the geometry in the moving coordinate systems. Therefore, it is very natural to subtract the term $\nabla \psi \cdot \mathbf{V}_0$ from the material derivative $\psi$ to define the shape derivative $\psi'$ with respect to the geometry in the stationary coordinates. If $\psi(\Omega)$ is independent on the geometric object $\Omega$, then we have $\psi'(\Omega; \mathbf{V}) = 0$.

Furthermore, the definition of shape derivative for a function $\varphi(S)$ defined over a two-dimensional manifold $S$, ensures that the shape derivative shows no dependence on the extension of $\varphi$ in the near neighbourhood. We propose the following proposition to show that the first variation of a functional on $S$ is closely related to the shape derivative.

**Proposition 2.2.** Let $S$ be a two-dimensional smooth manifold in $\bar{D}$ with smooth boundary $\Gamma$, and $\mathbf{V}$ be a speed vector field such that $\mathbf{V} \in C(C^k(\bar{D}, \bar{D}); [0, \epsilon_0])$. Suppose that $\varphi = \varphi(S)$ is such given that the material derivative $\dot{\varphi}(S; \mathbf{V})$ and the shape derivative $\varphi'(S; \mathbf{V})$ exist. Then, the shape functional $J(S) = \int_S \varphi(S) dS$ is shape differentiable and we have

$$
\delta J(S; \mathbf{V}) = \int_S \varphi'(S; \mathbf{V}) dS + \int_S \varphi(S) H \mathbf{V}_0 \cdot \mathbf{n} dS + \int_{\Gamma} \varphi(S) \mathbf{V}_0 \cdot \mathbf{c}_r d\Gamma,
$$

where $H$ is the mean curvature of the surface $S$, and $\mathbf{c}_r$ is the unit co-normal vector. Furthermore, if $\varphi(S) = \psi(\Omega)\big|_S$, then we have

$$
\delta J(S; \mathbf{V}) = \int_S \psi'(\Omega; \mathbf{V})\big|_S dS + \int_S \left(\frac{\partial \psi}{\partial \mathbf{n}} + \psi H\right) \mathbf{V}_0 \cdot \mathbf{n} dS + \int_{\Gamma} \psi \mathbf{V}_0 \cdot \mathbf{c}_r d\Gamma.
$$

(2.27)
Proof. According to the change of variables $x = T_\epsilon(V)(X)$, and by using the transformation $T_\epsilon(V)$, the shape functional $J(S_\epsilon)$ over the perturbed surfaces $S_\epsilon$ can be expressed as follows:

\[(2.28)\quad J(S_\epsilon) = \int_{S_\epsilon} \varphi(S_\epsilon) \, dS_\epsilon = \int_S (\varphi \circ T_\epsilon(V))(X) \omega(X, \epsilon) \, dS,
\]

where $\omega(X, \epsilon)$ is defined as

\[(2.29)\quad \omega(X, \epsilon) = \det(DT_\epsilon(V)) \left| \frac{\partial T_\epsilon}{\partial X_j}(X) \right|_{i,j=1,2,3}.
\]

Note that the following expressions hold according to the Lemma 2.49 in Page 80 of [49],

\[(2.30)\quad \omega(X, 0) = 1, \quad \frac{\partial \omega(X, \epsilon)}{\partial \epsilon} \bigg|_{\epsilon=0} = \nabla_S \cdot V_0.
\]

Therefore, based on the definition of the first variation, we have

\[(2.31)\quad \delta J(S; V) = \lim_{\epsilon \to 0} \frac{J(S_\epsilon) - J(S)}{\epsilon} = \int_S \varphi(S_\epsilon) \circ T_\epsilon(V) \omega(X, \epsilon) - \varphi \circ T_0(V) \omega(X, 0) \, dS
\]

By using the integration by parts and also making use of Eq. (2.21), we obtain

\[(2.32)\quad \delta J(S; V) = \int_S \varphi(S; V_0) \, dS - \int_S \nabla_S \varphi(S) \cdot V_0 \, dS + \int_S \varphi(S) \mathcal{H} V_0 \cdot n \, dS
\]

Furthermore, if we assume that $\psi$ is a function defined on the domain $\Omega$, such that its restriction on $S$ is equal to the function $\varphi(S)$, namely

\[(2.33)\quad \psi(\Omega) \big|_S = \varphi(S),
\]

then, by noticing the difference of the definition of shape derivatives on the domain $\Omega$ and the surface $S$ (i.e., Eq. (2.20) and Eq. (2.21)), we can reformulate Eq. (2.32) in terms of the extension function $\psi(\Omega)$ as

\[\delta J(S; V) = \int_S \varphi'(S; V) \, dS + \int_S \left( \frac{\partial \varphi}{\partial n} + \psi \mathcal{H} \right) V_0 \cdot n \, dS + \int_\Gamma \psi V_0 \cdot c_\Gamma \, d\Gamma,
\]

which completes the proof. \qed
Remark 2.3. If $S$ is a closed surface, then the boundary term about $\Gamma$ in (2.27) will vanish. The similar results for a closed curve or surface can be found in [12, 21].

In the following, we will apply (2.27) in Proposition 2.2 for calculating the first variation of the energy (or shape) functional defined in (2.3), where the integrand is the surface energy density $\gamma(n)$. To calculate the shape derivatives and obtain the first variation, we shall make use of the signed distance function, which is a powerful tool in shape sensitivity analysis. Consider a closed domain $\Omega \subset \mathbb{R}^3$ with a smooth boundary surface $\partial \Omega$, and then the signed distance function is defined as

$$b(x) = \begin{cases} d(x, \partial \Omega), & \forall x \in \mathbb{R}^3 \setminus \Omega, \\ 0, & \forall x \in \partial \Omega, \\ -d(x, \partial \Omega), & \forall x \in \Omega. \end{cases}$$

(2.34)

Here, $d(x, \partial \Omega) = \inf_{y \in \partial \Omega} ||x - y||$. The signed distance function $b(x)$ can be used to determine the unit outer normal vector $n$ and the mean curvature $H$ on the boundary surface $\partial \Omega$. More precisely, we can extend the functions $n$ and $H$ which are defined on $\partial \Omega$ in terms of $b(x)$ in a tubular neighbourhood such that

$$n(x) = \nabla b(x) \bigg|_{\partial \Omega}, \quad H(x) = \Delta b(x) \bigg|_{\partial \Omega}, \quad \forall x \in \partial \Omega.$$  

(2.35)

The shape derivative of the signed distance function in the direction of a vector field $V$ is calculated as $b'(\Omega; V) = -V_0 \cdot n$ (see [12, 21] for more details). Moreover, based on the extension, the shape derivatives of the two extension functions restricted on $\partial \Omega$ are also obtained (see Lemma 3.1 in [12]), i.e.,

$$n'(\Omega; V) \bigg|_{\partial \Omega} = -\nabla_s (V_0 \cdot n), \quad H'(\Omega; V) \bigg|_{\partial \Omega} = -\Delta_s (V_0 \cdot n).$$

2.3. First variation. By applying Eq. (2.27) and making use of the shape derivative of the unit outer normal vector, we obtain the following lemma.

Lemma 2.1. Assume that $S \subset D$ is a two-dimensional smooth manifold with smooth boundary $\Gamma$. Let $n$ be the unit outer normal vector of $S$, and $V$ be a speed vector field such that $V \in C^\infty(\bar{D} \setminus D; [0, r_0])$. If the shape functional $J(S) = \int_S \gamma(n) \, ds$ with a surface energy (density) $\gamma(n)$, then the first variation of $J(S)$ is given as

$$\delta J(S; V) = \int_S \left( \nabla_s \cdot \xi \right) (V_0 \cdot n) \, ds + \int_{\Gamma} V_0 \cdot c^\gamma \, d\Gamma,$$

(2.37)

where $\xi := \xi(n)$ is the Cahn-Hoffman vector, which is defined previously in Eq. (1.2), and $V_0 \cdot n$ represents the deformation velocity along the outer normal direction of the interface $S$, and the vector $c^\gamma := (\xi \cdot n) c^\gamma - (\xi \cdot c^\gamma) n$ with $c^\gamma$ representing the unit co-normal vector (shown in Fig. 4.1).

Proof. We firstly assume $\gamma(p)$ is a homogeneous extension of $\gamma(n)$,

$$\gamma(p) = |p| \gamma \left( \frac{p}{|p|} \right), \quad \forall p \in \mathbb{R}^3 \setminus \{0\},$$

(2.38)

where the definition domain of the function $\gamma(n)$ changes from unit vectors $n$ to arbitrary non-zero vectors $p \in \mathbb{R}^3$.

We next consider a bounded domain $\Omega \subset \mathbb{R}^3$ such that $S \subset \partial \Omega$. Then, based on the signed distance function defined in (2.34), we can define $\nabla b(x) \in \mathbb{R}^3$ as an extension of the normal vector $n$ in the neighbourhood of $S$. Thus we can reformulate

$$J(S) = \int_S \gamma(\nabla b(x)) \, ds = \int_S \psi(\Omega) \, ds,$$

(2.39)
Finally, by combining the above three terms together, we immediately have
\begin{equation}
\psi'(\Omega; \mathbf{V})\big|_S = \nabla \dot{\gamma}(\nabla b(x)) \cdot \mathbf{n}'(\Omega; \mathbf{V})\big|_S = -\xi \cdot \nabla_s (V_0 \cdot \mathbf{n}).
\end{equation}

Moreover, by noting the fact \(|\nabla b(x)| = 1\), we obtain
\begin{equation}
\frac{\partial \psi}{\partial \mathbf{n}}\big|_S = \xi \cdot \left((\nabla \nabla b(x)) \cdot \nabla b(x)\right)\big|_S = 0,
\end{equation}
where \(\nabla \nabla b(x) \in \mathbb{R}^{3 \times 3}\). By making use of Eq. (2.27) and combining Eq. (2.40) and (2.41), we immediately have
\begin{equation}
\delta J(S; \mathbf{V}) = -\int_S \xi \cdot \nabla_s (V_0 \cdot \mathbf{n}) \, dS + \int_S \gamma(\mathbf{n}) (V_0 \cdot \mathbf{n}) \, \mathcal{H} \, dS + \int_{\Gamma} \gamma(\mathbf{n}) (V_0 \cdot \mathbf{c}_r) \, d\Gamma
\end{equation}
\begin{equation}
:= I + II + III.
\end{equation}

For the first term, by using the integration by parts, we obtain
\begin{equation}
I = \int_S (\nabla_s \cdot \xi) (V_0 \cdot \mathbf{n}) \, dS - \int_S (\xi \cdot \mathbf{n}) (V_0 \cdot \mathbf{n}) \, \mathcal{H} \, dS - \int_{\Gamma} (\xi \cdot \mathbf{c}_r) (V_0 \cdot \mathbf{n}) \, d\Gamma.
\end{equation}

Based on Eq. (1.3), we have \(\gamma(\mathbf{n}) = \xi \cdot \mathbf{n}\). Thus we can rewrite
\begin{equation}
II = \int_S (\xi \cdot \mathbf{n}) (V_0 \cdot \mathbf{n}) \, \mathcal{H} \, dS,
\end{equation}
\begin{equation}
III = \int_{\Gamma} (\xi \cdot \mathbf{n}) (V_0 \cdot \mathbf{c}_r) \, d\Gamma.
\end{equation}

Finally, by combining the above three terms together, we immediately have
\begin{equation}
\delta J(S; \mathbf{V}) = \int_S (\nabla_s \cdot \xi) (V_0 \cdot \mathbf{n}) \, dS + \int_{\Gamma} \left[ (\xi \cdot \mathbf{n}) \mathbf{c}_r - (\xi \cdot \mathbf{c}_r) \mathbf{n} \right] \cdot V_0 \, d\Gamma
\end{equation}
\begin{equation}
= \int_S (\nabla_s \cdot \xi) (V_0 \cdot \mathbf{n}) \, dS + \int_{\Gamma} \mathbf{c}_r \cdot V_0 \, d\Gamma,
\end{equation}
with \(\mathbf{c}_r = (\xi \cdot \mathbf{n}) \mathbf{c}_r - (\xi \cdot \mathbf{c}_r) \mathbf{n}\).

By using the above Lemma, we can easily obtain the first variation of the energy functional for solid-state dewetting problems defined in (2.3).

**Theorem 2.1.** The first variation of the free energy (or shape) functional (2.3) used in solid-state dewetting problems with respect to a smooth vector field \(\mathbf{V}\) can be written as:
\begin{equation}
\delta W(S; \mathbf{V}) = \int_S (\nabla_s \cdot \xi) (V_0 \cdot \mathbf{n}) \, dS + \int_{\Gamma} (\mathbf{c}_r \cdot \mathbf{n}_r + \gamma_{FS} - \gamma_{VS}) (V_0 \cdot \mathbf{n}_r) \, d\Gamma,
\end{equation}
where \(\mathbf{n}_r\) is the unit outer normal of the contact line curve \(\Gamma\) on the substrate (shown in Fig. 2.4).

**Proof.** From (2.3), we observe that the total free energy consists of two parts: the film/vapor interface energy \(W_{\text{int}}\) and the substrate energy \(W_{\text{sub}}\). First, by using Lemma 2.1, we can directly obtain the first variation of the film/vapor interface energy \(W_{\text{int}}\) as follows,
\begin{equation}
\delta W_{\text{int}}(S; \mathbf{V}) = \int_S (\nabla_s \cdot \xi) (V_0 \cdot \mathbf{n}) \, dS + \int_{\Gamma} V_0 \cdot \mathbf{c}_r \, d\Gamma.
\end{equation}
Here, $c_i^γ$ is a linear combination of $c_r$ and $n$, which is defined on the contact line $Γ$. Therefore, as shown in Fig. 2.1, we have

\[(2.49)\]

\[c_r \perp τ_r, \quad n \perp τ_r \Rightarrow c_i^γ \perp τ_r.\]

For solid-state dewetting problems studied in this paper, we assume that the contact line $Γ$ must move along the substrate plane $S_{sub}$, i.e.,

\[(2.50)\]

\[T_εΓ \subset S_{sub}, \quad \mathbf{V}_0(x) \parallel S_{sub}, \quad ∀x ∈ Γ.\]

Therefore, for any $x ∈ Γ$, $\mathbf{V}_0(x)$ can be decomposed into two vectors along the directions $n_r(x)$ and $τ_r(x)$, i.e., $\mathbf{V}_0 = k_1 n_r + k_2 τ_r$, where $k_1$ and $k_2$ represent the corresponding components. By making use of (2.49), we can obtain

\[\mathbf{V}_0 \cdot c_i^γ = (k_1 n_r + k_2 τ_r) \cdot c_i^γ = k_1 (n_r \cdot c_i^γ)\]

\[= (\mathbf{V}_0 \cdot n_r) (c_i^γ \cdot n_r), \quad ∀x ∈ Γ.\]

Thus we can reformulate (2.48) as

\[(2.51)\]

\[δW_{int}(S; V) = \int_S (\nabla S \cdot ξ) (\mathbf{V}_0 \cdot n) dS + \int_Γ (c_i^γ \cdot n_r)(\mathbf{V}_0 \cdot n_r) dΓ.\]

On the other hand, we can rewrite the substrate energy $W_{sub}$ as

\[(2.52)\]

\[W_{sub} = (γ_{FS} - γ_{VS})A(Γ) = (γ_{FS} - γ_{VS})\int_{S_{FS}} dS_{FS}.\]

By using Proposition 2.2 and noting that the integrand $ϕ$ in (2.26) is a constant and $S_{FS}$ is a flat surface with a plane boundary curve $Γ$ (i.e., $H = 0$ and $n_r$ is the unit co-normal vector of the flat surface $S_{FS}$), we directly have

\[(2.53)\]

\[δW_{sub}(S; V) = (γ_{FS} - γ_{VS})\int_Γ \mathbf{V}_0 \cdot n_r dΓ.\]

By combining Eqs. (2.51) and (2.53), we obtain the following conclusion

\[(2.54)\]

\[δW(S; V) = \int_S (\nabla S \cdot ξ) (\mathbf{V}_0 \cdot n) dS + \int_Γ (c_i^γ \cdot n_r + γ_{FS} - γ_{VS})(\mathbf{V}_0 \cdot n_r) dΓ,\]

which completes the proof. \(\square\)

Remark 2.4. The variational result given by (2.47) tells us that the rate of change of the total interfacial free energy is contributed from the two parts: one part results from the change of the interface $S$, and it is proportional to the weighted mean curvature (i.e., $\nabla S \cdot ξ$) \[\frac{1}{2} \frac{d}{dS} \] and the rate of change of the volume (i.e., $\mathbf{V}_0 \cdot n dS$, the normal velocity times the surface area element); the other part comes from the change of the contact line $Γ$.

Remark 2.5. In 2D case, the variational result given by (2.47) in Theorem 2.1 will reduce to the variational result presented in the reference [2].

Remark 2.6. When the substrate is curved in 3D, the variational result given by (2.47) in Theorem 2.1 is still valid. We can perform similar discussions as the reference [2] for curved substrates in 2D.
3. Equilibrium shapes. The equilibrium shape of the solid-state dewetting problem can be stated as follows [4, 25]:

\[
\min_{\Omega} W := W(S) = \int_S \gamma(n) \, dS + (\gamma_{FS} - \gamma_{VS}) A(\Gamma) \quad \text{s.t.} \quad |\Omega| = C,
\]

where \( C > 0 \) is a prescribed constant representing the total volume of the dewetted particle, and \( \Omega \) represents the domain (or the particle) enclosed by the interface \( S \) and the substrate plane \( S_{\text{sub}} \).

The Lagrangian for the above optimization problem can be defined as

\[
L(S, \lambda) = \int_S \gamma(n) \, dS + (\gamma_{FS} - \gamma_{VS}) A(\Gamma) - \lambda (|\Omega| - C),
\]

with \( \lambda \) representing the Lagrange multiplier. The first variation of the total volume term can be obtained by simply choosing the integrand \( \psi(x) \equiv 1, \forall x \in \Omega \) in (2.22) by Proposition 2.1. Therefore, by combining with Eq. (2.47), the first variation of the Lagrangian with respect to a smooth vector field \( V \) can be given as

\[
\delta L(S, \lambda; V) = \int_S (\nabla_S \cdot \xi - \lambda)(V_0 \cdot n) \, dS + \int_{\Gamma} (c^\gamma_{\Gamma} \cdot n_{\Gamma} + \gamma_{FS} - \gamma_{VS})(V_0 \cdot n_{\Gamma}) \, d\Gamma.
\]

Based on the above first variation, we have the following theorem which yields the necessary conditions for the equilibrium shape of solid-state dewetting problem.

**Theorem 3.1.** Assume that a two-dimensional manifold \( S_e \) with smooth boundary \( \Gamma_e \) is the equilibrium shape of the solid-state dewetting problem (3.1), then the following conditions must be satisfied

\[
\nabla_{S_e} \cdot \xi = \lambda, \quad \text{on} \quad S_e,
\]

\[
c^\gamma_{\Gamma} \cdot n_{\Gamma} + \gamma_{FS} - \gamma_{VS} = 0, \quad \text{on} \quad \Gamma_e.
\]

where the constant \( \lambda \) is determined by the prescribed total volume, i.e., the constant \( C \).

**Proof.** If \( S_e \) is the equilibrium shape, then (3.3) must vanish at \( S = S_e \) for any smooth vector field \( V \). Therefore, we immediately obtain the above two necessary conditions. \( \Box \)

For isotropic surface energy, i.e., \( \gamma(n) \equiv 1 \) (scaled by a constant \( \gamma_0 \)), we have \( \xi = n \) and \( c^\gamma_{\Gamma} = c_\Gamma \). By simple calculations, Eq. (3.4a) will be reduced to the condition of constant mean curvature. Denote \( \Gamma_e \) as the boundary of \( S_e \), for arbitrary \( x \in \Gamma_e \), let \( \theta_i(x) \) represent the equilibrium contact angle at boundary point \( x \). Then, Eq. (3.4b) will reduce to

\[
\cos \theta_i(x) = \sigma, \quad \forall x \in \Gamma_e,
\]

where the (dimensionless) material constant \( \sigma := \frac{\gamma_{FS} - \gamma_{VS}}{\gamma_0} = \cos \theta_i \), and it is the well-known isotropic Young equation [64].

Eq. (3.4b) can be regarded as the Young equation for anisotropic surface energy \( \gamma(n) \) in 3D. For the anisotropic case, we can write the surface energy density in terms of the spherical coordinate, i.e., \( \gamma_{FV} = \gamma(\theta, \phi) \) (scaled by a constant \( \gamma_0 \)). Therefore, the Cahn-Hoffman \( \xi \)-vector can be decomposed into the following three components (as shown in Fig. 3.1(b)):

\[
\xi(n) = \nabla \tilde{\gamma}(n) = \gamma(\theta, \phi) n + \frac{\partial \gamma(\theta, \phi)}{\partial \theta} \tau_\theta + \frac{1}{\sin \theta} \frac{\partial \gamma(\theta, \phi)}{\partial \phi} \tau_\phi,
\]
where in these expressions,

\[ \mathbf{n} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)^T, \]
\[ \mathbf{\tau}_\theta = (\cos \theta \cos \phi, \cos \theta \sin \phi, -\sin \theta)^T, \]
\[ \mathbf{\tau}_\phi = (-\sin \phi, \cos \phi, 0)^T. \]

Therefore, we obtain that the following expressions hold

\[ \mathbf{\xi} \cdot \mathbf{n} = \gamma(\theta, \phi), \quad \mathbf{c}_r \cdot \mathbf{n}_r = \cos \theta(\mathbf{x}). \]
\[ \mathbf{\xi} \cdot \mathbf{c}_r = \frac{\partial \gamma(\theta, \phi)}{\partial \theta}, \quad \mathbf{n} \cdot \mathbf{n}_r = \sin \theta(\mathbf{x}). \]

Thus we can rewrite Eq. (3.4b) as

\[ \gamma(\theta, \phi) \cos \theta(\mathbf{x}) - \frac{\partial \gamma(\theta, \phi)}{\partial \theta} \sin \theta(\mathbf{x}) - \sigma = 0, \quad \forall \mathbf{x} \in \Gamma_e, \]

which is consistent with the anisotropic Young equation discussed for the solid-state dewetting problem in 2D [4, 55].

If \( \mathbf{X} := \mathbf{X}(\theta, \phi) \) represents the position vector of a surface, we have \( \nabla_S \cdot \mathbf{X} = 2 \) by using Definition 2.1. Therefore, if we use the \( \mathbf{\xi} \)-plot to represent the position vector of the equilibrium shape, then the necessary condition (3.4a) will be automatically satisfied. From one side, this is the reason why the \( \mathbf{\xi} \)-plot can yield equilibrium shapes for a free-standing solid particles (as shown in Fig 1.1). Furthermore, based on the recent work for the generalized Winterbottom construction [4, 57], we can construct its analytical expression for the equilibrium shape which also can satisfy the contact angle condition (3.4b). First, we define a domain of definition \( U_\phi \) for \( \theta \) under a fixed value \( \phi \) as

\[ U_\phi := \left\{ \theta \big| \gamma(\theta, \phi) \cos \theta - \frac{\partial \gamma(\theta, \phi)}{\partial \theta} \sin \theta - \sigma \geq 0, \quad \theta \in [0, \pi] \right\}, \]

where \( \sigma = \frac{\gamma_{VS} - \gamma_{FS}}{\gamma_0} \). Based on Theorem 3.1 we can explicitly construct its equilib-
rium shape in the parametric formula as $S_e(\theta, \phi) := X(\theta, \phi) = (x(\theta, \phi), y(\theta, \phi), z(\theta, \phi))^T$, where

$$\begin{align*}
x(\theta, \phi) &= \lambda \left[ \gamma(\theta, \phi) \sin \theta \cos \phi + \frac{\partial \gamma(\theta, \phi)}{\partial \theta} \cos \theta \cos \phi - \frac{1}{\sin \theta} \frac{\partial \gamma(\theta, \phi)}{\partial \phi} \sin \phi \right], \\
y(\theta, \phi) &= \lambda \left[ \gamma(\theta, \phi) \sin \theta \sin \phi + \frac{\partial \gamma(\theta, \phi)}{\partial \theta} \cos \theta \sin \phi + \frac{1}{\sin \theta} \frac{\partial \gamma(\theta, \phi)}{\partial \phi} \cos \phi \right], \\
z(\theta, \phi) &= \lambda \left[ \gamma(\theta, \phi) \cos \theta - \frac{\partial \gamma(\theta, \phi)}{\partial \theta} \sin \theta - \sigma \right],
\end{align*}$$

where $\phi \in [0, 2\pi]$, $\theta \in U_{\phi}$, and $\lambda$ is the scaling constant determined by the total volume $|\Omega|$.

$$\begin{align*}
\text{(a)} & \quad \text{(b)} & \quad \text{(c)} \\
\text{(d)} & \quad \text{(e)} & \quad \text{(f)}
\end{align*}$$

Fig. 3.2. The equilibrium shape defined by Eq. (3.11), where (a)-(c) is for isotropic surface energy, i.e., $\gamma(\mathbf{n}) \equiv 1$, but with different material constants $\sigma = \cos(\pi/3), \cos(\pi/2), \cos(3\pi/4)$, respectively; (d) $\gamma(\mathbf{n}) = 1 + 0.2(n_1^4 + n_2^4 + n_3^4)$, $\sigma = \cos(3\pi/4)$; (e) The surface energy density is given by Eq. (1.4), where $\sigma = \cos(3\pi/4)$, $\varepsilon = 0.01$; (f) The surface energy density is given by $\gamma(M_x(\pi/4)\mathbf{n})$ where $\gamma(\mathbf{n})$ is defined by Eq. (1.4) and $M_x(\pi/4)$ represents an orthogonal matrix for the rotation by an angle $\pi/4$ about the $x$-axis in 3D, using the right-hand rule, where $\sigma = \cos(3\pi/4)$, $\varepsilon = 0.01$.

Based on the formula (3.11), the equilibrium shape under different types of surface energy anisotropies, e.g., the cubic anisotropy and regularized “cusped” anisotropy defined in Eq. (1.4), can be easily constructed. Fig. 3.2(a)-(c) depicts the equilibrium shapes for isotropic surface energy with the material constant $\sigma$ chosen as $\sigma = \cos(\pi/3), \cos(\pi/2), \cos(3\pi/4)$, respectively. It clearly demonstrates the effect of the material constant $\sigma$ on the equilibrium shape by influencing the equilibrium contact angle via Eq. (3.5). Moreover, we also present equilibrium shapes for the cubic anisotropic surface energy, i.e., $\gamma(\mathbf{n}) = 1 + a(n_1^4 + n_2^4 + n_3^4)$ and regularized “cusped” surface energy defined in Eq. (1.4) with $\sigma = \cos(3\pi/4)$ in Fig. 3.2(d)-(e). The anisotropy for Fig. 3.2(f) is chosen by an anti-clockwise rotation along the $x$-axis by 45 degrees under the right-hand rule for the regularized “cusped” surface energy. We can observe that this rotation results in a corresponding rotation of the equilibrium shape.

4. A sharp-interface model and its properties. In this section, we propose a kinetic sharp-interface model for simulating solid-state dewetting of thin films with anisotropic surface energies, and then we show that the proposed model satisfies the mass conservation and energy dissipation.

4.1. The model. Based on Eq. (2.47) in Theorem 2.1 we can define the first variation of the total interfacial energy functional with respect to the film/vapor
interface \( S \) and its boundary curve (i.e., the contact line \( \Gamma \)) as

\[
\frac{\delta W}{\delta S} = \nabla_S \cdot \xi, \quad \frac{\delta W}{\delta \Gamma} = c^r \cdot \mathbf{n}_r + \gamma_{FS} - \gamma_{VS}.
\]

From the Gibbs-Thomson relation \([38, 52]\), the chemical potential can be defined as

\[
\mu = \Omega_0 \frac{\delta W}{\delta S} = \Omega_0 \nabla_S \cdot \xi,
\]

with \( \Omega_0 \) representing the atomic volume. The normal velocity of the moving interface is controlled by surface diffusion \([7, 38, 55, 25]\), and it can be defined as follows by Fick’s laws of diffusion \([3]\)

\[
J = -D_s \nu k_B T_e \nabla_S \mu, \quad v_n = -\Omega_0 (\nabla_S \cdot J) = D_s \nu \Omega_0 \frac{k_B T_e}{\Omega_0} \nabla^2 S \mu.
\]

In these expressions, \( J \) is the mass flux of atoms, \( D_s \) is the surface diffusivity, \( k_B T_e \) is the thermal energy, \( \nu \) is the number of diffusing atoms per unit area, \( \nabla_S \) is the surface gradient. In addition to the surface diffusion which controlled the motion of the moving interface, we still need the boundary condition for the moving contact line. Following the idea for simulating solid-state dewetting in 2D \([55, 25, 24]\), we assume that the normal velocity of the contact line \( \Gamma \) is simply given by the energy gradient flow, which is determined by the time-dependent Ginzburg-Landau kinetic equations, i.e.,

\[
v_n = -\eta \frac{\delta W}{\delta \Gamma} = -\eta (c^r \cdot \mathbf{n}_r + \gamma_{FS} - \gamma_{VS}),
\]

with \( 0 < \eta < \infty \) denoting the contact line mobility, which can be thought of as a reciprocal of a constant friction coefficient. For the physical explanation behind this approach, please refer to the recent paper \([55]\).

We choose the characteristic length scale and characteristic surface energy scale as \( h_0 \) and \( \gamma_0 \), respectively, the time scale as \( h_0^2 B_0 \gamma_0 \) with \( B = \frac{k_B T_e}{D_s \nu \Omega_0} \), and the contact line mobility is scaled by \( B h_0^3 \). Let \( X(\cdot) = (x(\cdot), y(\cdot), z(\cdot))^T \) be a local parameterization of the moving film/vapor interface \( S \), then we can obtain a dimensionless kinetic sharp-interface model for solid-state dewetting of thin film via the following Cahn-Hoffman \( \xi \)-vector formulation as

\[
\begin{align}
\partial_t X &= \Delta_S \mu \mathbf{n}, \quad t > 0, \\
\mu &= \nabla_S \cdot \xi, \quad \xi(\mathbf{n}) = \nabla^2 \gamma(p)|_{p=n},
\end{align}
\]

where \( t \) is the time, \( \mathbf{n} \) is the unit outer normal vector of \( S \), and \( \xi := \xi(\mathbf{n}) \) is the Cahn-Hoffman vector (scaled by \( \gamma_0 \)). Here, for simplicity, we still use the same notations for all the dimensionless variables.

Let \( X(\cdot, t) = (x(\cdot, t), y(\cdot, t), z(\cdot, t))^T \) represents a parameterization of the moving contact line \( \Gamma(t) \). The initial condition is given as \( S_0 \) with boundary \( \Gamma_0 \) such that

\[
S_0 := X(\cdot, 0) = (x_0, y_0, z_0), \quad \Gamma_0 := X(\cdot, 0)|_{\Gamma}.
\]

The above governing equations are subject to the following boundary conditions:
(i) contact line condition
\begin{equation}
   z_{\Gamma}(\cdot,t) = 0, \quad t \geq 0;
\end{equation}

(ii) relaxed contact angle condition
\begin{equation}
   \partial_t X_{\Gamma} = -\eta \left( c_{\Gamma}^* \cdot n_{\Gamma} - \sigma \right) n_{\Gamma}, \quad t \geq 0;
\end{equation}

(iii) zero-mass flux condition
\begin{equation}
   \left( c_{\Gamma} \cdot \nabla_S \mu \right)_{|\Gamma} = 0, \quad t \geq 0;
\end{equation}

where \( \eta \) represents a (dimensionless) contact line mobility, \( c_{\Gamma}^* \) is the anisotropic co-normal vector which is defined as \( c_{\Gamma}^* := (\xi \cdot n) c_{\Gamma} - (\xi \cdot c_{\Gamma}) n \), \( c_{\Gamma} \) represents the co-normal vector, and \( n_{\Gamma} = (n_{\Gamma,1}, n_{\Gamma,2}, 0)^T \) is the outer unit normal vector of \( \Gamma \) on the flat substrate (cf. Fig. 2.1), and \( \sigma = \frac{\gamma_{0,S} - \gamma_{0,F}}{\gamma_{0}} \) is a (dimensionless) material constant.

**Remark 4.1.** For isotropic surface energy, i.e., \( \gamma(n) \equiv 1 \), we obtain that \( \xi = n \) and \( \mu = \nabla \gamma \cdot \xi = \nabla \gamma \cdot n = H \); for anisotropic surface energy, by Definition 2.1 and some calculations, we can obtain that the dimensionless chemical potential \( \mu \) is the weighted mean curvature discussed in [53].

**Remark 4.2.** The contact line condition in Eq. (4.8) ensures that the contact line must move along the substrate plane. Because the contact line \( \Gamma \) lies on the substrate (i.e., \( Oxy \) plane), the third component of \( n_{\Gamma} \) is always zero, i.e., \( n_{\Gamma,3} = 0 \). As long as the initial condition satisfies \( z_{\Gamma}(\cdot,0) = 0 \), it can automatically satisfy the boundary condition (i) \( z_{\Gamma}(\cdot,t) = 0, \forall t > 0 \) by using the boundary condition (ii). The last boundary condition (iii) ensures that the total volume/mass of the thin film is conserved during the evolution, i.e., no-mass flux at the moving contact line.

**Remark 4.3.** The above governing equation is well-posed when the surface energy is isotropic or weakly anisotropic. But when the surface energy is strongly anisotropic, some missing orientations will appear on equilibrium shapes [47, 50]; in this case, the governing equation becomes ill-posed, and it can be regularized by adding regularization terms such that the regularized sharp-interface model is well-posed [25, 6]. For the analytical criteria about the classification of surface energy anisotropy in 3D, interested readers could refer to [47].

### 4.2. Mass conservation and energy dissipation.

In the following, we will rigorously prove that the proposed sharp-interface model satisfies the mass conservation and the total free energy dissipation during the evolution.

**Proposition 4.1.** Assume that \( X(\cdot,t) \) is the solution of the sharp-interface model, i.e., Eqs. (4.5) - (4.6) with boundary conditions (4.8) - (4.10), and denote \( S(t) := X(\cdot,t) \) as the moving film/vapor interface. Then, the total volume (or mass) of the thin film, labeled as \( |\Omega(t)| \), is conserved, i.e.,
\begin{equation}
   |\Omega(t)| \equiv |\Omega(0)|, \quad t \geq 0.
\end{equation}

Furthermore, the (dimensionless) total interfacial free energy of the system is non-increasing during the evolution, i.e.,
\begin{equation}
   W(t) \leq W(t_1) \leq W(0) = \int_{S(0)} \gamma(n) \, dS - \sigma A(\Gamma(0)), \quad t \geq t_1 \geq 0.
\end{equation}
Proof. By making use of the first variation (2.22) and simply choosing the integrand \( \psi(x) \equiv 1, \forall x \in \Omega \), and using the governing equation (4.5), we can calculate the time derivative of the total volume as (noting that \( V_0 = \partial_t \mathbf{X} \))

\[
\frac{d}{dt} |\Omega(t)| = \int_{S(t)} \partial_t \mathbf{X} \cdot \mathbf{n} \, dS = \int_{S(t)} \Delta_s \mu \, dS = 0, \quad t \geq 0,
\]

where the last equality comes from the integration by parts and the zero-mass flux condition (4.10), and it indicates that the total volume/mass is conserved.

To obtain the time derivative of the (dimensionless) total free energy, by making use of Theorem 2.1 and Eq. (2.47), but replacing the perturbation variable \( \epsilon \) with the time variable \( t \), we can immediately obtain

\[
\frac{d}{dt} W(t) = \int_{S(t)} (\nabla_{\mathbf{s}} \cdot \mathbf{\xi}) (\partial_t \mathbf{X} \cdot \mathbf{n}) \, dS + \int_{\Gamma(t)} (\mathbf{c}_r^\gamma \cdot \mathbf{n}_r - \sigma) (\partial_t \mathbf{X}_\Gamma \cdot \mathbf{n}_r) \, d\Gamma.
\]

By substituting the governing equations and the relaxed contact angle boundary condition, i.e.,

\[
\mu = \nabla_{\mathbf{s}} \cdot \mathbf{\xi}, \quad \Delta_s \mu = \partial_t \mathbf{X} \cdot \mathbf{n}, \quad \partial_t \mathbf{X}_\Gamma \cdot \mathbf{n}_r = -\eta (\mathbf{c}_r^\gamma \cdot \mathbf{n}_r - \sigma),
\]

into the above equation and using the integration by parts and the zero-mass flux condition, we obtain

\[
\frac{d}{dt} W(t) = \int_{S(t)} \mu \Delta_s \mu \, dS - \eta \int_{\Gamma(t)} (\mathbf{c}_r^\gamma \cdot \mathbf{n}_r - \sigma)^2 \, d\Gamma
\]

\[
- \int_{S(t)} |\nabla_{\mathbf{s}} \mu|^2 \, dS - \eta \int_{\Gamma(t)} (\mathbf{c}_r^\gamma \cdot \mathbf{n}_r - \sigma)^2 \, d\Gamma \leq 0, \quad t \geq 0,
\]

where the constant \( \eta > 0 \). The last inequality immediately implies the energy dissipation. \( \square \)

Remark 4.4. In the above proof, we need to calculate the time derivatives of the total volume and the total free energy. These two derivatives can be easily obtained by making use of the speed method and the first variation presented in Section 2. In Section 2, we consider any type of smooth perturbations. In fact, a family of evolving interface surfaces \( \{S(t)\}_{t \geq 0} \) can be also thought of as a type of perturbations, only by replacing the perturbation variable \( \epsilon \) with the time variable \( t \). Therefore, the time derivatives can be directly obtained by using the first variation of the total volume functional and the total free energy functional.

5. Numerical results. In this section, we perform numerical simulations for solid-state dewetting in 3D to investigate the morphological evolution of thin films in various cases. We implement the parametric finite element method (PFEM) \( \square \) for solving the proposed sharp-interface model in 3D. For the detailed introduction of numerical algorithms about PFEM in 3D, interested readers could refer to \( \square \).

5.1. Equilibrium convergence. We have presented a mathematical description of the equilibrium shape in Section 3. Here, we present some numerical convergence results to equilibrium shapes by numerically solving the proposed kinetic sharp-interface model.

From the relaxed contact angle boundary condition (4.9), which describes the migration of the contact line, we know that the contact line mobility \( \eta \) precisely...
controls the relaxation rate of the contact angle towards its equilibrium state. The large $\eta$ will accelerate the relaxation process \[^{55}\]. Here, we numerically investigate the effect of $\eta$ on the evolution of the dynamic contact angle. The evolution surfaces \(\{S(t_m)\}_{m=1}^{M}\) are discretized by polygonal surfaces such that \(S^m = \bigcup_{j=1}^{N} D_{j}^m\), where \(\{D_{j}^m\}_{j=1}^{N}\) are mutually disjoint triangle surfaces, and the polygonal surface \(S^m\) has $K$ vertices given as \(\{q_{km}^{m}\}_{k=1}^{K}\). The boundary of the polygonal surface \(S^m\) given by polygonal $\Gamma^m = \bigcup_{j=1}^{Ne} h_{j}^m$, where \(\{h_{j}^m\}_{j=1}^{Ne}\) are line segments of the curve ordered in counter-clockwise direction when viewing from the top. We define the following average contact angle as the indicator for convergence,

\[
\bar{\theta}^m = \frac{1}{N_c} \sum_{j=1}^{N_c} \arccos(\mathbf{c}_{r_{m},j} \cdot \mathbf{n}_{r_{m},j}),
\]

where $\mathbf{c}_{r_{m},j}$ and $\mathbf{n}_{r_{m},j}$ are the unit vectors defined on the $j$-th line segment $h_{j}^m$ of the contact line curve. Fig. 5.1 shows the temporal evolution of $\bar{\theta}^m$ and the normalized energy $W(t)/W(0)$ under different choices of the contact line mobility $\eta$. The initial thin film chosen as a unit cube. From the figure, we can observe that the larger mobility $\eta$ will accelerate the process of relaxation such that the contact angles evolve faster towards its equilibrium contact angle $3\pi/4$. As shown in Fig. 5.1, the energy decays faster for larger mobility, but finally the equilibrium contact angle converges to the same value. It indicates that the equilibrium contact angles as well as the equilibrium shape are independent of the choice of the contact line mobility $\eta$. In the following numerical simulations, the contact line mobility is chosen to be very large (e.g., $\eta = 100$). This choice of $\eta$ will result in a very quick convergence to the equilibrium contact angle (defined by Eq. (3.4b)). The detailed investigation of the influence of the parameter $\eta$ on the solid-state dewetting evolution process and equilibrium shapes was performed in 2D \[^{55}\].

We next show a convergence result between the numerical equilibrium shape by solving the proposed sharp-interface model and its theoretical equilibrium shape. Fig. 5.2 depicts equilibrium shapes under different mesh sizes, where $\sigma = \cos(15\pi/36)$,
Fig. 5.2. Comparisons of the cross-section profiles along the x-direction of the numerical equilibrium shapes under different meshes with its theoretical equilibrium shape, where the initial shape is chosen as a (1, 2, 1) cuboid, the surface energy $\gamma(n) = 1 + 0.25(n_1^4 + n_2^4 + n_3^4)$, and $\sigma = \cos(15\pi/36)$.

$\gamma(n) = 1 + 0.25(n_1^4 + n_2^4 + n_3^4)$. The initial shape is chosen as a (1, 2, 1) cuboid, then we numerically evolve it until the equilibrium state by using different meshes, which are given by a set of small isosceles right triangles. If we define the mesh size indicator $h$ as the length of the hypotenuse of the isosceles right triangle, then “Mesh 1” represents the initial mesh with $h = h_0 = 0.125$, and the time step is chosen as $\tau = \tau_0 = 0.00125$ for numerical computation. Meanwhile, the time step for “Mesh 2” ($h = h_0^2$) and “Mesh 3” ($h = h_0^4$) are chosen as $\tau = \frac{\tau_0}{4}$ and $\tau = \frac{\tau_0}{16}$, respectively. For a better comparison, we plot the cross-section profiles along the x-direction for the numerical equilibrium shapes and the theoretical equilibrium shape. As shown in Fig. 5.2, we can clearly observe that as the computational mesh size gradually decreases, the numerical equilibrium shapes uniformly converge to the theoretical equilibrium shape (constructed by Eq. (3.11)).

5.2. Kinetic evolution. First, we focus on the case for isotropic surface energy, i.e., $\gamma(n) \equiv 1$. We start with numerical examples for an initially, short cuboid island with (2, 2, 1) representing its length, width and height, respectively (as shown in Fig. 5.3(a)). The computational parameter is chosen as $\sigma = \cos \frac{5\pi}{6}$. As can be seen in Fig. 5.3, we show several snapshots of the morphology evolution for the short cuboid towards its equilibrium shape. As time evolves, the initial sharp corners and edges on the island become smooth in a very short time (Fig. 5.3(b)), and finally the island film forms a spherical shape as its equilibrium shape (Fig. 5.3(f)).

Short cuboid island films tend to form a single spherical island shape as its equilibrium shape minimizing the total free energy (i.e., the minimal surface area). However, the morphological evolution for long cuboid islands could be quite different. Due to the Plateau-Rayleigh instability [31, 45, 36], long cuboid islands could pinch off and break up into a number of small isolated particles on the substrate before they form a single spherical shape as its equilibrium. In order to investigate this phenomenon, we perform the simulation by fixing the same material constant as $\sigma = \cos(3\pi/4)$, and choosing the shape of initial island film as a long cuboid with (1, 12, 1). For isotropic case, as can be seen in Fig. 5.4, the island quickly evolves into a cylinder-like shape during the evolution; then it accumulates more and more materials near the two edges, while its neck becomes thinner and thinner; finally, it pinches off at the neck.
Fig. 5.3. Several snapshots during the evolution of an initially, cuboid island film with isotropic surface energy towards its equilibrium shape: (a) $t = 0.0$; (b) $t = 0.1$; (c) $t = 0.2$; (d) $t = 0.5$; (e) $t = 0.7$; (f) $t = 1.40$, where the initial shape of the thin film is chosen as a $(2, 2, 1)$ cuboid, and the material constant is chosen as $\sigma = \cos(5\pi/6)$.

Fig. 5.4. Several snapshots during the evolution of an initial, cuboid island film with isotropic surface energy until its pinch-off time: (a) $t = 0$; (b) $t = 0.01$; (c) $t = 0.30$; (d) $t = 0.50$; (e) $t = 0.80$; (f) $t = 1.03$, where the initial shape is chosen as a $(1, 12, 1)$ cuboid, and the material constant $\sigma = \cos(3\pi/4)$.

and breaks up into two small isolated islands on the substrate. For cubic anisotropic surface energies, long cuboid islands also exhibit the similar pinch-off process as the isotropic surface energy case. We test the numerical example for an initially cuboid island with the same material constant and initial shape, as shown Fig. 5.5. From the figure, we observe that it finally forms three isolated small islands, while it only produces two isolated small islands in the isotropic surface energy case. This indicates that for this type of cubic anisotropic surface energy, the solid island tends to dewet more easily than in the isotropic surface energy.
Fig. 5.5. Several snapshots during the evolution of an initial, cuboid island film with anisotropic surface energy until its pinch-off time: (a) $t = 0$; (b) $t = 0.020$; (c) $t = 0.10$; (d) $t = 0.24$; (e) $t = 0.54$; (f) $t = 0.695$, where the initial shape is chosen as a $(1, 1, 2)$ cuboid, the material constant $\sigma = \cos(3\pi/4)$, and the anisotropic surface energy is chosen as the cubic type, i.e., $\gamma(n) = 1 + a(n_1^4 + n_2^4 + n_3^4)$ with $a = 0.25$.

Fig. 5.6. Several snapshots during the evolution of an initial, cuboid island film with isotropic surface energy towards its equilibrium shape: (a) $t = 0$; (b) $t = 0.004$; (c) $t = 0.008$; (d) $t = 0.0120$; (e) $t = 0.020$; (f) $t = 0.080$, where the initial shape is chosen as a $(3.2, 3.2, 0.1)$ cuboid, and the material constant $\sigma = \cos(5\pi/6)$.

Finally, we investigate the morphological evolution of square island films with size $(m, m, h)$ on a flat substrate. We start by simulating the evolution of an initial, small square island with size $(3.2, 3.2, 0.1)$, and the material constant is chosen as $\sigma = \cos(5\pi/6)$. As can be seen in Fig. 5.6, the four corners of the square island retract much more slowly than the middle points of the four edges at the beginning, thus resulting in an almost cross shape for the island film (see Fig. 5.6(d)). This phenomenon about “mass accumulation” at the corner has also been observed in experiments [54, 61, 63] or numerical simulations by a phase-field approach [23, 39]. Subsequently, because the square island is small, these retracting corners eventually catch up with the edges, and
Fig. 5.7. Several snapshots during the evolution of an initial, cuboid island film with isotropic surface energy until its pinch-off time: (a) $t = 0$; (b) $t = 0.005$; (c) $t = 0.010$; (d) $t = 0.031$, where the initial shape is chosen as a $(6.4, 6.4, 0.1)$ cuboid, and the material constant $\sigma = \cos(5\pi/6)$.

Fig. 5.8. The cross-section profile of the island film along its y-direction and diagonal direction for the example shown in Fig. 5.7: (a) $t = 0$; (b) $t = 0.005$; (c) $t = 0.010$; (d) $t = 0.031$.

The contact line begins to move towards a circular shape in order to form a spherical shape as its equilibrium. During the evolution, we can also observe that a valley forms at the center of the island, but finally it disappears. To observe the possible pinch-off phenomenon, we enlarge the square size and simulate the evolution of an initial, large square island with size $(6.4, 6.4, 0.1)$ (shown in Fig. 5.7). From the figure, we observe that the valley at the center becomes deeper and deeper, and it eventually touches the substrate, and produce a hole in the center of the island. We stop the numerical simulation at the moment when there exists one new mesh point which touches the substrate. For a better illustration, in Fig. 5.8, we also plot several snapshots about its corresponding cross-section profile of the island film during the evolution.

6. Conclusions. We proposed a sharp-interface approach for simulating solid-state dewetting of thin films in three dimensions (3D), and this approach can handle with the effect of the surface energy anisotropy. Based on the Cahn-Hoffman $\xi$-vector formulation and the speed method, we derived rigorously the first variation of the total free energy functional of the solid-state dewetting problem. From the first variation,
necessary conditions for the equilibrium shape of solid-state dewetting were rigorously given in mathematics. Furthermore, a kinetic sharp-interface model was also proposed for simulating the solid-state dewetting of thin films in 3D. The governing equations described the interface evolution which is controlled by surface diffusion and contact line migration. A lot of numerical examples were performed for solving the sharp-interface model, and numerical results reproduced the complex features in the solid thin film dewetting observed in experiments, such as edge retraction, hole formation, faceting, corner accumulation, pinch-off and Rayleigh instability.

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