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Some Problems in Adhesive Micro-contact Mechanics

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**A thesis submitted to the Nanyang Technological University
in fulfillment of the requirements for the degree of
Doctor of Philosophy**

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Summary

Adhesion between solid surfaces can induce contact between the surfaces. The fundamental mechanical issues in micro-scale differ from those in the macro world. In the present thesis, three fundamental adhesive micro-contact issues in liquid bridging and wafer bonding are studied.

Evolution of liquid bridge: The evolution process of a liquid bridge between a rigid cylinder and a rigid plane is investigated by setting up a theoretical model based on the framework of irreversible non-equilibrium thermodynamics. A weak statement is applied to the thermodynamics ingredients, leading to the Galerkin method in numerical analysis. Results from the numerical analysis describe the evolution history. The effects of the control parameters on the evolution process are discussed. Our numerical results show that the liquid droplets between contact surfaces would evolve to an equilibrium liquid bridge.

Liquid-bridging induced micro-contact: With a finite element method (FEM) simulation, the micro-scale contact induced by the liquid bridging is studied to have a much more in-depth understanding of the coupling effect between the solid deformation and the liquid bridging. A rigorous description of the solid surface under deformation is achieved. It is noticed that the contact due to the liquid bridging force and deformation of the elastic solid is coupled in the finite element analysis (FEA) simulation. Based on our coupled FEA numerical results, the accuracies of the decoupled and coupled Hertz approximation solutions are discussed. Under certain conditions, the errors of the Hertz approximate solutions increase dramatically.

Energy rate consideration in wafer bonding: An energy absorption rate criterion is proposed in analyzing the process of wafer bonding. We demonstrate it by a numerical simulation for the bonding process between a rigid plane and an elastic-plastic wafer with a periodical wavy surface. Wafer bonding depends on the competition between the work of adhesion of the contact interface and the resistance to

bonding. Wafer bonding can be divided into self-activated complete bonding, self-activated partial bonding, and external loading assisted bonding. The effects of a far field pressure, plasticity and asperity shape are also discussed.

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Chapter 2 A Global Analysis of Thermodynamics Evolution of Non-equilibrium Liquid Bridge

R	radius of the rigid cylinder
θ_1, θ_2	contact angles at two TPLs
ψ	filling angle
γ_{ij}	interface tensions between domains i and j
A_{ij}	interface areas between domains i and j
Δp	Laplace pressure (capillary pressure)
κ	local mean curvature of the meniscus.
l	half width of the liquid bridge area
G	free energy
$\delta \mathbf{r}_n$	virtual motion of the meniscus
$\delta \mathbf{r}_n^{(t1)}, \delta \mathbf{r}_n^{(t2)}$	virtual motions of two TPLs
R_1, R_2	first and second principal curvature radii of the meniscus
$\bar{n}$	unit vector normal to the meniscus
$\bar{n}^{(t1)}, \bar{n}^{(t2)}$	unit vectors normal to two TPLs respectively
f	thermodynamics force on the meniscus
f_{t1}, f_{t2}	thermodynamics forces along two TPLs
v_n	actual normal velocity of the meniscus
$v_n^{(t1)}, v_n^{(t2)}$	actual normal velocities of two TPLs
M	mobility of the meniscus
M_{t1}, M_{t2}	mobilities of two TPLs
κ_e	mean curvature of the equilibrium meniscus
$\{q_0, q_1, \dots, q_m\}$	generalized coordinates
$\{\dot{q}_0, \dot{q}_1, \dots, \dot{q}_m\}$	generalized velocities
$\Phi_i (i=1, \dots, m)$	shape function for the meniscus

$\Phi_i^{(t1)}, \Phi_i^{(t2)} (i=1, \dots, m)$	shape functions for two TPLs
$T_i (i = 1, \dots, m)$	generalized forces
H_{ij}	viscosity matrix
ψ_0	initial average filling angle
λ	wavelength of the liquid bridge
t_0	characteristic time
$\tilde{M}$	dimensionless mobility
$\tilde{\lambda}$	dimensionless wavelength of the liquid bridge
$\tilde{H}_{ij}$	dimensionless viscosity matrix
$\psi_{\min}, \psi_{\text{aver}}, \psi_{\max}$	minimum, average and maximum filling angles
$G0$	free energy of the system without liquid bridge
t_{eq}	equilibrium time
$\tilde{t}_{eq}$	normalized equilibrium time

Chapter 3 Elastic Analysis for Liquid-Bridging Induced Contact

ψ	filling angle
χ	a dimensionless parameter measuring the rigidity of the solid
E	elastics modulus of the half space
ν	Poisson ratio of the half space
R_0	radius of the rigid sphere
γ	surface tension of the liquid/air interface
Δp	Laplace pressure across the liquid/vapor interface
$2H$	mean curvature of the meniscus
θ_1, θ_2	contact angles at the two triple points
l	outer radius of the half space/liquid interface
r_1	first principal radius (meridian radius)
r_2	second principal radius
F_s	surface tension force
F_{sz}	axial component of surface tension force

F_{sr}	radial component of surface tension force
$F_{\Delta p}$	normal component of the capillary force
F_{total}	total normal force in the axial direction
c	contact radius
E^*	composite modulus
$\mathcal{C}_0$	initial contact radius
l_0	initial outer radius of liquid/half space interface

Chapter 4 Energy Rate Consideration in Wafer Bonding

Γ	the work of adhesion
γ_1, γ_2	surface tensions of two wafer materials
γ_{12}	interface tension
A	interface area
U	strain energy including plastic work installed in the solids
W	work generated by external loading
R	resistance (energy absorption rate)
E	Young's modulus of the wafer
ν	Poisson's ratio of the wafer.
E^*	effective modulus of the wafer
σ_s	yielding stress of the wafer
λ	asperity wavelength on the wafer surface
Δ	asperity amplitude on the wafer surface
α	indicating the asperity profile shape
a	half contact width
p	far filed pressure
$\overline{U}$	average strain energy over a wavelength
E_b	specific effective bonding energy
G	energy release rate

Chapter 1

Introduction

1.1 Overview

Between contact surfaces, a liquid bridging forming around contacting asperities on the surfaces due to surface energy effects (Fig. 1.1) or the adhesion between solid surfaces (e.g. Van der Waals force) can induce attraction forces (Fig. 1.2). Adhesive micro-contacts due to these attraction forces exist in various micro-electro-mechanical systems (MEMS) [1]. However, the adhesive micro-contact issues have been hardly touched by traditional tribology study, because the fundamental mechanical issues in micro-scale differ from those in the macro world. It has been noticed that some concepts commonly used in macro-scale cannot be directly applied in the micro level. The following aspects are noticeable in our analysis.

Firstly, in MEMS operation, stresses induced by the external loading are necessarily small. The attraction forces between contact surfaces should assume a greater importance than in the macroscopic world. Published reports [2-4] confirmed these expectations.

Secondly, the size of a MEMS device is very small. Therefore, the forces related to the volume, such as gravitational forces, are negligible; but the microscopic forces related to the surfaces and the interactions between surfaces, such as the attraction forces between contact surfaces, can be significant and dominant factors (Fig. 1.2). Contact exists between the surfaces due to these attraction forces, even there is no external forces added [5-7].

Finally, every manufacturing process induces micro-geometric defects and irregularities at the surface of engineering parts, including MEMS elements. Real surfaces are rough with a large number of asperities and actually in contact at these asperities [1, 8-13]. These asperities play an important role in the operating behavior

of the surfaces. The real contact pressure is determined by the asperity shapes, and is much different from the macro-scale pressure distribution on the interface

Adhesive micro-contact is the most important factor determining the properties and lives of products in modern MEMS devices. In a wide range of engineering applications, especially in the MEMS device development and application, components are often used in a wet environment where water condenses around surface asperities [5, 8, 9, 11, 12, 14]. In some of the cases, a small amount of liquid, such as a liquid lubricant, is intentionally introduced between two solid surfaces. Due to surface energy effects, concave liquid bridge forms around contacting asperities. An attraction force arises including the Laplace pressure inside the liquid bridge and the surface tension in the meniscus [15, 16]. This is often called liquid bridging phenomenon, and this attraction force is often called liquid bridging force. When the external applied forces are negligible, the liquid bridging force becomes a dominant factor and makes a significant contribution to the asperity contact. It may produce very large contact stresses over small real contact area comparable to the material yield stress. Therefore, it may greatly influence the static friction, kinetic friction and wear properties of the contact interfaces [8, 11, 12].

Adhesive contact is not always considered as negative aspect in the contact. The wafer bonding used in MEMS is a technology based on the mechanism of the adhesive contact. Complex structures consisting of structured and plain silicon or other wafers are joined to one mechanically stable configuration. This technology gives electronic and opto-electronic devices of much superior performance. In these devices, mirror polished, flat and clean solids are attracted into contact at room temperature under the short-range intermolecular and inter-atomic attraction forces [6, 17-22]. However, there exist geometrical and physical mismatches between the two wafer surfaces [19, 22, 23]. Two wafers are actually in micro-contact at asperities. Resistance exists during surfaces joining to form an interface. Whether or not two wafers bond depends on the micro-contact state under the attraction forces [6].

Investigators from various fields have studied various aspects of the adhesive micro-contact issues in the liquid bridging phenomenon [5, 12, 14, 24-29] and the wafer bonding technology [21, 30-33]. However, many basic phenomena related to adhesive

micro-contact mechanics are still not fully understood. This situation motivated the author to enter this research area.

There are some existing research results for the micro-contact in the liquid bridging phenomenon, where by considering the solids as rigid bodies, the issues of fluid mechanics in equilibrium have been discussed, such as the volume of the liquid, the surface area and mean curvature of the meniscus, and the forces exerted on the solid surfaces [5, 12, 14, 24-29]. In terms of the well-known Hertzian contact solution for the contact between a sphere and a plane, the liquid-mediated contact in micro-tribology systems has been studied for the contact area and pressure on the contact interface. However, the liquid bridging force was just added into the normal force to estimate its contribution to the real contact area as well as the friction [34].

There are still some important issues in liquid bridging phenomena missed in the existing research works. The evolution process of a liquid bridge from a non-equilibrium initial configuration to an equilibrium state has not been investigated up to now. Moreover, in most of the existing works, the exact influence of the solid deformation on the meniscus shape and the liquid bridging force was not taken into account. It is suspected that the liquid bridging force and the solid deformation could be significantly coupled under certain conditions. In order to have a much more in-depth understanding of the contribution of the coupling effect, the influence of the solid deformation on the liquid bridging force should be researched more rigorously.

For the micro-contact in wafer bonding technology, large real contact area close or equal to full nominal contact area has been observed, where asperity contacts are coupled. The existing micro-contact mechanics (e.g., Johnson et al, 1971 [35]) mainly treats the micro-contact with small real contact area and the surface asperities contact are taken as independent single asperity contact. It cannot treat the micro-contact problems in wafer bonding technology. Therefore, many theoretical models have been proposed to explain the micro-scale mechanism of wafer bonding. The well known are the gap-closing model [21, 30-32] and Yu and Suo's model on complete bonding [33]. Their models were concentrated only on the complete bonding (bonding over the entire interface) and based on the balance between the strain energy installed in the elastic bodies and the released surface energy from the contact interface during the bonding

process. It cannot explain certain phenomena, such as partial bonding (bonding over part of the interface), external loading assisted bonding, and measurement of the bonding strength of the interface [6]. The micro-contact in wafer bonding technology needs further study.

1.2 Objective and scope

Based on the above overview of adhesive micro-contact mechanics existing in liquid bridging phenomenon and wafer bonding technology, the objective of the present research work are to study some fundamental micro-contact mechanics issues in liquid bridging phenomenon and wafer bonding technology. The following research progresses have been achieved:

(1) To investigate the time-dependent evolution process of the liquid bridge from an initial non-equilibrium state to its corresponding final static equilibrium state. By following an irreversible non-equilibrium thermodynamics framework for the micro-structural evolution of dynamical systems [36], a theoretical model was set up for the kinetic evolution process of a liquid bridge. The ingredients of the non-equilibrium thermodynamics for the liquid bridging induced contact between a solid cylinder and a rigid plane were considered and presented theoretically. Then, the weak statement was applied to these ingredients to set up a set of nonlinear equations for describing the dynamical system of the liquid bridging induced contact. Next, some assumptions were proposed for describing the shape of the liquid bridge in the evolution process. The dynamical evolution process of the liquid bridging was numerically simulated. Finally, based on the numerical results, the effects of the controlling parameters on the evolution process were discussed.

(2) The micro-scale contact induced by the liquid bridging force was studied to have a much more in-depth understanding of the contribution of the coupling effect between the solid deformation and the liquid bridging (i.e., the liquid bridging force and the meniscus shape). Attention was focused on the interaction between the size of the contact area and magnitude of the liquid-bridging force. The liquid-bridging force is

related to the contact area and other parameters, such as the filling angle, the contact angles, and the surface tension of the liquid by solving the Laplace-Young equation. When the liquid bridge exists between a rigid sphere and a half space, the solid deformation was obtained by finite element method (FEM). The surface profile of the elastically deformable half space was described accurately. Therefore, the influence of the solid deformation on the liquid bridging force and the meniscus shape was considered rigorously. Two approximate solutions for the same problem were also given and compared with the solutions of FEM. The errors inhabited in the approximate solutions were discussed because of ignoring the coupling effect. Parametric studies were carried out to reveal the dependence of the contact area and the liquid bridging force on the filling angle and the dimensionless group in the micro-scale contact.

(3) To investigate the phenomenon and micro-mechanism in the wafer bonding process due to the attraction force between solids with rough surfaces. A new criterion for wafer bonding, called energy absorption rate criterion in the present research work, will be proposed from the point of view of mechanics. According to this criterion, self-activated complete bonding, self-activated partial bonding, external loading assisted bonding and the bonding strength of the interface are discussed. For the demonstration purpose, we numerically analyzed a micro-contact exists between a rigid plane and an elastic-plastic wafer with a one-dimensional wavy sinusoidal surface where the strain energy and the resistance variation with the bonding area. Our newly proposed criterion is applied to the bonding process to describe the phenomenon in the entire process from partial bonding to complete bonding. The difference between the newly proposed criterion and the criterion proposed by former researchers is discussed.

1.3 Organization of the thesis

Chapter 2 describes the modeling and numerical implementation processes for the evolution of the non-equilibrium liquid bridging induced contact. Firstly, the

ingredients of the non-equilibrium thermodynamics for the liquid bridging induced contact between a solid cylinder and a rigid plane were given. They include the free energy of the system, the motions of the meniscus and the two triple point lines (TPLs), the driving force of the evolution, and the kinetic laws describing the relations between the actual velocities and the driving forces. Then, a weak statement, a terminology of variational calculus, combines the kinetic laws and the free energy variation associated with virtual motions of the meniscus and the two TPLs. It leads to the Galerkin method in numerical analysis of a set of nonlinear ordinary differentiation equations (ODEs) formulating the dynamical evolution process of the liquid bridging induced contact. Finally, the results from the numerical simulation processes were reported. The effects of the controlling parameters on the evolution process were discussed.

Chapter 3 describes the FEM numerical simulation process and results for the micro-scale contact between an elastically deformable half space and a rigid sphere induced by liquid bridging. Firstly, the general aspects of the liquid bridging induced contact were introduced. The more rigorous description of the half space surface under deformation was emphasized. Secondly, the coupled and decoupled Hertz contact solutions were also presented. Thirdly, a dimensional analysis was reported to reveal that all the three solutions are determined by two governing parameters. Next, the variations of the liquid bridging force and the contact area from FEM numerical simulations were presented. Finally, the numerical solution was compared with the Hertz approximate solutions and the coupling effect between the solid deformation and the liquid bridge is then revealed.

Based on energy rate concept, Chapter 4 reports a newly proposed wafer-bonding criterion. It was demonstratively applied to the bonding process between a rigid plane and a wafer with a one-dimensional wavy sinusoidal surface. Firstly, the general concept for this new criterion for predicting wafer bonding process is reported. Then, by FEM numerical simulation, the variations of the strain energy and the resistance with the real contact area are presented for the bonding process between a rigid plane and a wafer with a one-dimensional wavy sinusoidal surface. Finally, our newly proposed criterion is applied to analyze the bonding process and to reveal the

phenomena happening in the bonding process. The analysis results are compared with those from former criteria. Chapter 5 summarizes the main findings of this research.

Fig. 1.1 A liquid bridging forming between contact surfaces

(a) Schematic for a rough surface in contact with a flat surface with a liquid bridging; (b) schematic of contact area and meniscus area in a contacting asperity.

Fig. 1.2 Micro-scale forces versus surface separation

(where F_{lb} is the liquid bridging force, F_{el} the electro-static force and F_{vdw} the Van der Waals force)

Chapter 2

A Global Analysis of Thermodynamics Evolution of Non-equilibrium Liquid Bridge

2.1 Introduction

Real surfaces are rough with a large number of asperities. In a wide range of engineering applications, especially in the MEMS device development and application, components are often used in a wet environment where water condensate around surface asperities. In some of the cases, a small amount of liquid, such as a liquid lubricant, is intentionally introduced between two solid surfaces [5, 8, 9, 11, 12, 14].

With the presence of a thin liquid film or a droplet of liquid at the contact interface, concave liquid bridge forms around contacting asperities due to surface energy effects. An attraction force arises including the Laplace pressure inside the liquid bridge and the surface tension in the meniscus [15, 16]. This is often called liquid bridging phenomenon, and this attraction force is often called liquid bridging force.

Adhesive micro-contact induced by liquid bridging force is the most important factor determining the properties and lives of products in modern MEMS devices. When the external applied forces are negligible, the liquid bridging force becomes a dominant factor and makes a significant contribution to the asperity contact. It may produce very large contact stresses over small real contact area comparable to the material yield stress or / and hardness. Therefore, it may greatly influence the static friction, kinetic friction and wear properties of the contact interfaces [8, 9, 11, 12, 37, 43, 44].

Investigators from various fields have attacked various aspects of the liquid-bridging phenomenon. Of importance in applications are the volume of liquid, the area and mean curvature of the meniscus, the force exerted on the solid surfaces, and the contact area and pressure on the contact interface. Orr et al [5] presented a detailed literature

reviews on the earlier works related to the meniscus properties and capillary force induced by liquid bridging phenomenon between two solid surfaces. They also discussed the general case of a sphere contacting a plate with zero separation by solving the Laplace-Young's equation (see Finn, 1986 [25]; Bhushan, 1999 [12]). Gao et al [14] considered a general meridian meniscus profile by the solution of second-order partial differential equations derived from Pascal's law for incompressible liquids. Their study showed that the liquid-bridging force formula derived from surface energy consideration with the assumption of spherical curvature is still applicable, as long as the lateral size of the liquid bridge is much smaller than the radii of the spherical asperity tips. Lian et al [26] considered the liquid-bridging force between two rigid spheres. All these works mainly discussed the issues of fluid mechanics for liquid bridge in equilibrium between two solid surfaces. Having noticed that the earlier studies did not address the coupling effect between the deformation of solids and liquid bridging (i.e., the solids were considered as rigid bodies), Fan and Gao [38] analyzed a coupled liquid bridging induced contact problem under two-dimensional plane strain condition, and obtained a solution by applying the singular integral equation approach.

However, all the studies mentioned above discussed only the liquid bridging phenomenon in equilibrium. The evolution process of the liquid bridge from a non-equilibrium initial configuration to an equilibrium state has not been investigated up to now. Therefore, in the present work, we study on the entire time-dependent evolution process starting from a non-equilibrium liquid bridge.

Following the irreversible non-equilibrium thermodynamics framework for the micro-structural evolution of dynamical systems [36], we propose a theoretical model focused on the kinetic evolution process of the liquid bridge from its non-equilibrium initial configuration toward the equilibrium state. In terms of the global approach to micro-structural evolution in materials [36, 39, 40], we describe the processes of meniscus evolution and triple-point-line (TPL) motion. The model aims to describe the entire process of the kinetic evolution, not only to predict the final equilibrium state of the liquid bridge, but also to record the entire history of the evolution process.

2.2 Liquid-bridging induced contact at equilibrium state

Let us consider a two-dimensional plane strain contact problem, where an infinitely long rigid cylinder laying on the surface of a rigid half space. A small liquid drop is introduced between the surfaces of the cylinder and the half space. The liquid wets two solid surfaces due to the surface energy effect and a liquid bridge forms. The intersection normal to the axis of the cylinder is shown in Fig. 2.1 (Figures are to be found at the end of the chapter), where $Oxyz$ is a right-handed Cartesian coordinate system, with y -axis parallel to the cylinder axis, x -axis on the surface of the half space, and z -axis on the symmetric plane. R is the radius of the rigid cylinder, θ_1 and θ_2 the equilibrium contact angles at two TPLs that the meniscus makes with the rigid cylinder and the rigid half space respectively; ψ is called the filling angle and is a measure of the liquid volume. The equilibrium configuration of this system is independent of y , so that the problem is considered as “plane strain”. The whole system is divided into 4 domains by the cylinder surface, the flat surface of the half space, and the meniscus, which are marked as domains 1, 2, 3, and 4, respectively in Fig. 2.1. Therefore, γ_{12} , γ_{13} , γ_{24} , γ_{34} , and γ_{14} are the interface tensions of the liquid/cylinder, liquid/plane, cylinder/vapor, plane/vapor, and liquid/vapor (i.e., the meniscus) interfaces, respectively. For this equilibrium system, the contact angles are related to the surface tensions by Young’s equations,

$$\begin{aligned}\gamma_{24} &= \gamma_{12} + \gamma_{14} \cos \theta_1 \\ \gamma_{34} &= \gamma_{13} + \gamma_{14} \cos \theta_2\end{aligned}\tag{2.1}$$

It is clear that the equilibrium contact angles are system constants as the interface tensions are physical properties.

The pressure difference across the meniscus (commonly referred to as Laplace pressure or capillary pressure) is determined by Laplace equation,

$$\Delta p = 2H\gamma_{14} ,\tag{2.2}$$

where $2H$ is the local mean curvature of the meniscus.

When the liquid volume is small, or the characteristic length in the system is small, the gravity effect of the liquid is ignorable. Therefore, the Laplace pressure and the mean curvature of the meniscus can be assumed to be constant in the entire liquid and the on the entire meniscus respectively. Following Orr et al's analysis [5], the meridian meniscus profile at equilibrium state is approximated by an arc of circle with a radius R_1 in Fig. 2.2. Through a geometrical analysis, the radius of this arc of circle is found to be

$$R_1 = \frac{R(1 - \cos \psi)}{\cos(\psi + \theta_1) + \cos \theta_2} . \quad (2.3)$$

As the problem is of plane strain, the mean curvature of the meniscus is

$$\kappa \equiv 2H = 1/R_1 . \quad (2.4)$$

The half width of the liquid bridge area, as shown in Fig. 2.2, is given as

$$l = R \sin \psi + R_1 \cdot [\sin(\psi + \theta_1) - \sin \theta_2] . \quad (2.5)$$

The liquid-bridging force per unit length of the cylinder is given by

$$F = l \cdot \Delta p + \gamma_{14} \sin \theta_2 . \quad (2.6)$$

The system described by Eq. (2.1) ~ Eq. (2.6) is at equilibrium state.

2.3 General consideration for non-equilibrium thermodynamic evolution of liquid bridge

Let us consider the contact problem between a rigid cylinder and a rigid half space with droplets of liquid present at the contact interface. At the beginning, the system is far from the equilibrium state. Due to surface energy effect, the liquid wets the two solids and the non-equilibrium liquid bridge forms. Different from the equilibrium configuration, the intersection of the liquid bridge normal to the cylinder axis varies. The problem is no longer of "plane strain". The whole system is still divided into the 4

domains as shown in Fig. 2.1. In Fig. 2.2, the meniscus is described by an equation $x = f(y, z)$. The filling angle ψ and the half width of the liquid bridge area are only functions of y . Driven by the reduction in the free energy of the system, the system evolves towards its final equilibrium configuration. In the present work, we set up a theoretical model based on the framework of non-equilibrium thermodynamics for the evolution of this liquid bridge [36, 39]. The evolution process and the final equilibrium state of the liquid bridge are predicted. In this section, many thermodynamics ingredients are considered, such as the free energy, driving force, and the kinetics of the system.

2.3.1 Gibbs free energy of the system

The liquid bridge evolution toward the equilibrium state is driven by the change of free energy, G . The free energy of the system consists of an internal energy G_0 (the energy of the material system in its reference frame) and a potential energy Π imposed to the system by the Laplace pressure:

$$G = G_0 + \Pi . \quad (2.7)$$

For the system considered here, the internal energy comes from the interface energies among different domains:

$$G_0 = \iint \gamma dA = \gamma_{12}A_{12} + \gamma_{13}A_{13} + \gamma_{24}A_{24} + \gamma_{34}A_{34} + \gamma_{14}A_{14} , \quad (2.8)$$

where γ_{ij} and A_{ij} are the specific interface tensions and the interface areas between domains i and j respectively. The interface tensions are isotropic and constant. The internal energy relates to the global quantities, i.e., the interface areas in the system. On the other hand, the potential energy is the negative work supply W imposed to the system by the Laplace pressure

$$\Pi = -W = -\iint_{A_{14}} t_i u_i dA , \quad (2.9)$$

where t_i is the i^{th} component of the Laplace pressure, and u_i the i^{th} component of the displacement of the meniscus. It is needed to point out that t_i and the meniscus area

A_{14} varies with the evolution process, and the Laplace pressure Δp is constant in the liquid at a given time.

Since Eqs. (2.8) and (2.9) only involve integration, the evaluations of the internal energy and potential energy are robust and insensitive to the local variation of microstructures. The free energy has a global feature. This leads to Galerkin method as shown in the following sections.

Non-equilibrium thermodynamics requires that the system evolve to decrease the free energy of the system.

2.3.2 Motion of meniscus and two TPLs; driving forces

Free energy by itself is insufficient to determine the shape change of the liquid bridge, because countless ways of shape change would reduce the free energy. To evolve the shape of the liquid bridge, the model needs more ingredients.

Fig. 2.3a and 2.3b illustrate the virtual motions of the meniscus and two TPLs, which resulting from the surface diffusion on the meniscus and the long-range mass diffusion in the liquid. A virtual motion of the meniscus is a small movement in the direction normal to the meniscus that need not obey any kinetic law. The amount of the motion, δr_n , can differ from point to point over the meniscus. Similarly, the virtual motions of two TPLs are small movements in the directions normal to the relevant TPL and on the solid surfaces. They also need not obey any kinetic law. The amount of the virtual motions of two TPLs, $\delta r_n^{(1)}$ and $\delta r_n^{(2)}$, can differ from point to point along the relevant TPL. All the virtual motions must conserve the mass of the system, i.e., the volume of the liquid must be conserved in the evolution process.

Despite of the form of the equation describing the meniscus, the virtual motions of two TPLs are found to be

$$\delta r_n^{(1)} = R \delta \psi / \sqrt{1 + (R \psi_{,y})^2} ; \delta r_n^{(2)} = \delta l / \sqrt{1 + (l_{,y})^2} \quad (2.10)$$

where, $\psi_{,y}$ and $l_{,y}$ denote the derivatives of ψ and l with respect to y .

At a given time, the meniscus is a smooth curved surface, and two TPLs are smooth curved lines. Here, the area of the surface elements on the meniscus is denoted by dA , the lengths of the line elements on TPLs 1 and 2 by ds_ψ and ds_l . The mean curvature of the meniscus is the sum of two principal curvatures

$$\kappa = 2H = \frac{1}{R_1} + \frac{1}{R_2} = \frac{(1+q^2)r - 2pqs + (1+p^2)t}{(1+p^2+q^2)^{3/2}}, \quad (2.11)$$

where R_1 and R_2 are two principal curvature radii of the meniscus, taken to be positive when the curvature center is outside the liquid. All the parameters in Eq. (2.11) are given as

$$r = \frac{\partial^2 x}{\partial y^2}, \quad s = \frac{\partial^2 x}{\partial y \partial z}, \quad t = \frac{\partial^2 x}{\partial z^2}, \quad p = \frac{\partial x}{\partial y}, \quad \text{and} \quad q = \frac{\partial x}{\partial z}, \quad (2.12)$$

where $x = f(y, z)$ is the equation describing the meniscus. However, we do not use the accurate value of κ , but an approximate value as in the following section.

Associated with the virtual motions δr_n , $\delta r_n^{(r1)}$, and $\delta r_n^{(r2)}$, the variations of all the interface areas are given as

$$\begin{aligned} \delta A_{14} &= -\iint_{A_{14}} \kappa \delta r_n dA + \int_{L_{r1}} \delta r_n^{(r1)} \cos \theta_1(y) ds_\psi + \int_{L_{r2}} \delta r_n^{(r2)} \cos \theta_2(y) ds_l \\ \delta A_{12} &= \int_{L_{r1}} \delta r_n^{(r1)} ds_\psi = -\delta A_{24} \\ \delta A_{13} &= \int_{L_{r2}} \delta r_n^{(r2)} ds_l = -\delta A_{34} \end{aligned} \quad (2.13)$$

and the volume of the liquid varies by

$$\delta V = \iint_{A_{14}} \delta r_n dA. \quad (2.14)$$

The above two integrals extend over the meniscus A_{14} and along two TPLs denoted by L_{r1} and L_{r2} . At a given time, the current contact angles at two TPLs are not constant and vary along the TPLs, i.e., they are functions of y , denoted by $\theta_1(y)$ and $\theta_2(y)$.

According to geometrical analysis, the unit vector normal to the meniscus directing to the vapor phase is given by

$$\vec{n} = (\vec{i} - x_y \vec{j} - x_z \vec{k}) / \sqrt{1 + (x_y)^2 + (x_z)^2}, \quad (2.15)$$

where $\vec{i}$, $\vec{j}$, and $\vec{k}$ are the unit vectors in the directions of x , y , and z axes, respectively.

The unit vector normal to the cylinder surface along L_{t1} is $\sin\psi\vec{i} - \cos\psi\vec{k}$. Therefore, at a given time, the current contact angle of the liquid along L_{t1} , $\theta_1(y)$, is the inclination angle between two vectors normal to the meniscus and the cylinder surface, which is given by

$$\begin{aligned} \cos\theta_1(y) &= \vec{n} \bullet (\sin\psi\vec{i} - \cos\psi\vec{k}) = (\sin\psi\vec{i} - \cos\psi\vec{k}) \bullet (\vec{i} - x_y \vec{j} - x_z \vec{k}) / \sqrt{1 + (x_y)^2 + (x_z)^2} \\ &= (\sin\psi + x_z \cos\psi) / \sqrt{1 + (x_y)^2 + (x_z)^2} \end{aligned} \quad (2.16)$$

Similarly, the current contact angle of the liquid along L_{t2} , $\theta_2(y)$, is given by

$$\cos\theta_2(y) = \vec{n} \bullet \vec{k} = \vec{k} \bullet (\vec{i} - x_y \vec{j} - x_z \vec{k}) / \sqrt{1 + (x_y)^2 + (x_z)^2} = -x_z / \sqrt{1 + (x_y)^2 + (x_z)^2} \quad (2.17)$$

During the evolution process of the liquid bridge, along the two TPLs, the virtual motions must satisfy the following contact conditions:

$$\delta r_n = \delta r_n^{(t1)} \sin\theta_1(y) \text{ along } L_{t1} \quad (2.18a)$$

$$\delta r_n = \delta r_n^{(t2)} \sin\theta_2(y) \text{ along } L_{t2} \quad (2.18b)$$

The volume of the liquid must be conserved. Therefore, we have

$$\delta V = \iint_{A_{t4}} \delta r_n dA = 0 \quad (2.19)$$

Associated with the virtual motions, the free energy of the system varies by δG as given by

$$\begin{aligned}
\delta G &= \delta G_0 - \delta W = \delta \left(\iint \gamma dA \right) + \iint_{A_{l4}} \Delta p \delta r_n dA \\
&= \int_{L_{r1}} \gamma_{12} \delta r_n^{(r1)} ds_\psi - \int_{L_{r1}} \gamma_{24} \delta r_n^{(r1)} ds_\psi + \int_{L_{r2}} \gamma_{13} \delta r_n^{(r2)} ds_l - \int_{L_{r2}} \gamma_{34} \delta r_n^{(r2)} ds_l \\
&\quad - \iint_{A_{l4}} \kappa \gamma_{14} \delta r_n dA + \int_{L_{r1}} \gamma_{14} \delta r_n^{(r1)} ds_\psi + \int_{L_{r2}} \gamma_{14} \delta r_n^{(r2)} ds_l + \iint_{A_{l4}} \Delta p \delta r_n dA
\end{aligned} \tag{2.20}$$

where the Laplace pressure, Δp , can be determined by the system force balance condition. However, as the Laplace pressure is constant at a given time and the liquid bridge volume is conserved, $\iint_{A_{l4}} \Delta p \delta r_n dA = 0$. Therefore, we need not calculate the value of Δp .

Define a thermodynamics force, f , as the free energy decrease associated with unit increased volume of the liquid on the meniscus, and two thermodynamics forces, f_{l1} and f_{l2} , as the free energy decrease associated with unit increased areas of the liquid/cylinder interface and the liquid/half space interface, namely

$$\int_{L_{r1}} f_{l1} \delta r_n^{(r1)} ds_\psi + \int_{L_{r2}} f_{l2} \delta r_n^{(r2)} ds_l + \iint_{A_{l4}} f \delta r_n dA = -\delta G \tag{2.21}$$

Since the virtual motions, δr_n , $\delta r_n^{(r1)}$, and $\delta r_n^{(r2)}$, are arbitrary functions of the positions on the meniscus and along two TPLs, respectively, the virtual motions, δr_n , $\delta r_n^{(r1)}$, and $\delta r_n^{(r2)}$, and Eq. (2.21) uniquely and completely define the quantities, f , f_{l1} and f_{l2} at every point on the meniscus and along the two TPLs, respectively. For the above-defined thermodynamics forces, f has a unit of pressure (force/area or energy/volume), and f_{l1} and f_{l2} have a unit of surface energy (force/length or energy/area). They are the driving forces of the thermodynamics evolution of the liquid bridge.

A comparison of Eqs. (2.20) with (2.21) gives the driving forces as

$$f_{l1} = \gamma_{24} - \gamma_{12} - \gamma_{14} \cos \theta_1(y) = \gamma_{14} [\cos \theta_1 - \cos \theta_1(y)] \tag{2.22a}$$

$$f_{l2} = \gamma_{34} - \gamma_{13} - \gamma_{14} \cos \theta_2(y) = \gamma_{14} [\cos \theta_2 - \cos \theta_2(y)] \tag{2.22b}$$

$$f = \kappa \gamma_{14} - \Delta p \tag{2.22c}$$

Eq. (2.22c) expresses the driving pressure on the meniscus, f , in terms of the current mean curvature of the meniscus, κ , the interface tension, γ_{14} , and the Laplace pressure, Δp . As expected, γ_{14} tends to drive the meniscus toward the curvature center, and Δp tends to pull the meniscus to the center of the liquid bridge. Eqs. (2.22a) and (2.22b) express the driving forces along the two TPLs, f_{t1} and f_{t2} , in terms of the current contact angles at two TPLs, $\theta_1(y)$ and $\theta_2(y)$, and the relevant interface tensions. They have a clear physical significance that the f_{t1} and f_{t2} are the sum of the interface tensions projected along the unit vectors normal to relevant TPLs in the surfaces in which the virtual motions, $\delta r_n^{(t1)}$ and $\delta r_n^{(t2)}$, are defined.

2.3.3 Kinetic laws

The energetics described in Sections 2.3.1 and 2.3.2 (the free energy and the thermodynamics forces) alone neither determine the evolution path, nor select the final state of a micro-structural evolution. The actual evolution path, and therefore the final equilibrium state, is determined by the energetics and kinetics, and they result from a compromise between the energetics and the kinetics. All kinetic processes induce dissipation. The kinetic laws characterize the relation between the driving forces and the material flows for the dissipation in a micro-structural evolution, the latter represented by both the current material structures (including various material flow amounts) and the material fluxes (i.e., the actual velocities of the meniscus and two TPLs in this evolution problem).

Let v_n be the actual normal velocity of the meniscus directing to vapor phase, $v_n^{(t1)}$ the actual normal velocity of L_{t1} on the cylinder surface directing from liquid phase to vapor phase, and $v_n^{(t2)}$ the actual normal velocity of L_{t2} on the half space surface directing from liquid phase to vapor phase, respectively. Following the framework of non-equilibrium thermodynamics for micro-structural evolution, these actual velocities are taken to be functions of the driving forces. Specifically, they are assumed to be linearly proportional to the driving forces as (i.e., linear kinetics)

$$v_n^{(t1)} = M_{t1} f_{t1} \quad (2.23a)$$

$$v_n^{(r2)} = M_{i2} f_{i2} \quad (2.23b)$$

$$v_n = M f \quad (2.23c)$$

Here M_{i1} , M_{i2} , and M are the mobility of the two TPLs and the meniscus, respectively. They will be used as phenomenological parameters of the model, to be determined by comparing model predictions with experimental observations. Thermodynamics requires that the meniscus and two TPLs move in the direction that reduces the free energy G of the system, so that all the three mobility parameters are positive.

Eqs. (2.23a, b, c) are the kinetic laws for motions of the meniscus and two TPLs. The actual motion of the system both conserves mass and obeys these kinetic laws.

Here it should be noted that the driving forces in Eqs. (2.23) are defined at every point on the meniscus and two TPLs. The kinetic laws, Eqs. (2.23), are local in that the velocity of a point only depends on the driving forces at the same point. Moreover, all the mobility parameters are assumed uniform and isotropic in the present analysis, thus they are material constants for our kinetic model. Therefore, the kinetic laws used in this work are linear relations. For a universal system, the kinetic laws may be extended to nonlinear and non-local relations [41].

The free energy, motion descriptions, driving forces and kinetic laws discussed above define the dynamics of the thermodynamics evolution. At a given time, the free energy variation corresponding to virtual motions determines the driving forces, and the kinetic laws update the liquid shape for a small time step. The process repeats for many time steps to evolve the system up to the equilibrium state.

2.3.4 Motion equation and equilibrium conditions

A combination of equations (2.22) and (2.23) leads to

$$v_n^{(r1)} = M_{i1} (\gamma_{24} - \gamma_{12} - \gamma_{14} \cos \theta_1(y)) = M_{i1} \gamma_{14} [\cos \theta_1 - \cos \theta_1(y)] \quad (2.24a)$$

$$v_n^{(r2)} = M_{i2} (\gamma_{34} - \gamma_{13} - \gamma_{14} \cos \theta_2(y)) = M_{i2} \gamma_{14} [\cos \theta_2 - \cos \theta_2(y)] \quad (2.24b)$$

$$v_n = M(\kappa\gamma_{14} - \Delta p) \quad (2.24c)$$

These partial differential equations govern the motions of the system. They are called motion equations for the system. The evolution process of the system depends on the solution of the motion equations under the contact conditions (2.18), volume conservation condition (2.19), and certain initial and boundary conditions.

As the kinetic laws, Eqs. (2.23), are local, the method to evolve the system by solving Eqs. (2.24) is a local approach [40, 42].

At the end of the evolution process, the system arrives at an equilibrium state, and the free energy of the system attains the minimum value. The system remains static, and the driving forces and the evolution velocities vanish. Setting the driving forces f, f_{t1} , and f_{t2} equal to 0, or setting the evolution velocities $v_n, v_n^{(r1)}$, and $v_n^{(r2)}$ equal to 0, leads to equilibrium conditions for the meniscus and two TPLs as

$$\gamma_{24} - \gamma_{12} - \gamma_{14} \cos \theta_1 = 0 \quad (2.25a)$$

$$\gamma_{34} - \gamma_{13} - \gamma_{14} \cos \theta_2 = 0 \quad (2.25b)$$

$$\Delta p = \kappa_e \gamma_{14} \quad (2.25c)$$

where θ_1, θ_2 and κ_e are the contact angles at two TPLs and the mean curvature of the meniscus when the system is at equilibrium state. This equation is same as Eqs. (2.1) and (2.2). Therefore, they are the well-known Laplace-Young's equations, which are frequently used to predict the shape of the liquid when a system with liquid bridging phenomenon is at static equilibrium state.

2.4 Weak statement and Galerkin method

It is quite formidable to solve the set of partial differential equations (2.24) in addition to certain conditions because of their nonlinear features. Moreover, solving Eqs. (2.24) is not a good way to look at the general problem for several reasons. Firstly,

Eqs. (2.24) are incorrect when interface tensions are anisotropic. Secondly, as a general problem often has to be analyzed approximately, a set of partial differential equations need not be a good starting point. Following Suo [36], the nonlinear dynamic system can be formulated by a weak statement, which circumvents the difficulties of anisotropy, and leads to the Galerkin method in numerical analysis. This method has been successfully used to model many non-equilibrium evolution problems [36, 39].

Replace the driving forces f, f_{t1} , and f_{t2} in Eqs. (2.21) with the corresponding velocities $v_n, v_n^{(r1)}$, and $v_n^{(r2)}$ of the meniscus and two TPLs by using the kinetic laws (2.23), giving

$$\int_{L_{t1}} \frac{v_n^{(r1)}}{M_{t1}} \delta r_n^{(r1)} ds_\psi + \int_{L_{t2}} \frac{v_n^{(r2)}}{M_{t2}} \delta r_n^{(r2)} ds_l + \iint_{A_{t4}} \frac{v_n}{M} \delta r_n dA = -\delta G(\delta r_n^{(r1)}, \delta r_n^{(r2)}, \delta r_n) \quad (2.26)$$

This is the weak statement for the dynamic system of the liquid bridge evolution by following the terminology of variational calculus. It means that the actual velocities $v_n, v_n^{(r1)}$, and $v_n^{(r2)}$ must satisfy Eq. (2.26) for virtual motions $\delta r_n, \delta r_n^{(r1)}$, and $\delta r_n^{(r2)}$ of arbitrary distribution on the meniscus and along the two TPLs.

In using the weak statement, it is often impractical to find the actual velocities for arbitrary virtual motions. Rather, one may find the approximate values of the actual velocities $v_n, v_n^{(r1)}$, and $v_n^{(r2)}$ for a restricted family of virtual motions. Obviously, the larger the family, the more accurate the approximation.

We can describe both the meniscus and the two TPLs with $m+1$ degrees of freedom, writing $\bar{q} = \{q_0, q_1, \dots, q_m\}$ for the generalized coordinates, and $\dot{\bar{q}} = \{\dot{q}_0, \dot{q}_1, \dots, \dot{q}_m\}$ for the corresponding generalized velocities. Based on the differential geometry, the evolving meniscus A_{t4} can be expressed by the position vector, $\bar{x} = \bar{x}(t)$, at a given time. Using the generalized coordinates, this position vector is expressed as $\bar{x} = \bar{x}(q_0, q_1, \dots, q_m)$, with the time t implicitly contained in the generalized coordinates. Similarly, the two evolving TPLs can be expressed by two position vectors $\bar{x}^{(r1)}$ and $\bar{x}^{(r2)}$ at a given time, which are expressed as $\bar{x}^{(r1)} = \bar{x}^{(r1)}(q_0, q_1, \dots, q_m)$ and

$\bar{x}^{(t2)} = \bar{x}^{(t2)}(q_0, q_1, \dots, q_m)$ respectively, also with the time t implicitly contained in the generalized coordinates. Therefore, the virtual motions δr_n , $\delta r_n^{(t1)}$ and $\delta r_n^{(t2)}$ are linear in the virtual variations of the generalized coordinates:

$$\delta r_n^{(t1)} = \sum_{i=0}^m \left(\bar{n}^{(t1)} \cdot \frac{\partial \bar{x}^{(t1)}}{\partial q_i} \right) \delta q_i \equiv \sum_{i=0}^m N_i^{(t1)} \delta q_i \quad (2.27a)$$

$$\delta r_n^{(t2)} = \sum_{i=0}^m \left(\bar{n}^{(t2)} \cdot \frac{\partial \bar{x}^{(t2)}}{\partial q_i} \right) \delta q_i \equiv \sum_{i=0}^m N_i^{(t2)} \delta q_i \quad (2.27b)$$

$$\delta r_n = \sum_{i=0}^m \left(\bar{n} \cdot \frac{\partial \bar{x}}{\partial q_i} \right) \delta q_i \equiv \sum_{i=0}^m N_i \delta q_i \quad (2.27c)$$

where $\bar{n}^{(t1)}$ is the unit vector normal to L_{t1} in the liquid/cylinder interface, $\bar{n}^{(t2)}$ the unit vector normal to L_{t2} in the liquid/half space interface, and $\bar{n}$ the unit vector normal to the meniscus directing to the vapor phase. $N_i^{(t1)}$, $N_i^{(t2)}$ and N_i ($i = 0, 1, \dots, m$) are the corresponding shape functions before applying the constraint condition of volume conservation, which depend on the generalized coordinates but not on the generalized velocities. Moreover, the actual velocities v_n , $v_n^{(t1)}$, and $v_n^{(t2)}$ are linear in the generalized velocities:

$$v_n^{(t1)} = \sum_{i=0}^m N_i^{(t1)} \dot{q}_i \quad (2.28a)$$

$$v_n^{(t2)} = \sum_{i=0}^m N_i^{(t2)} \dot{q}_i \quad (2.28b)$$

$$v_n = \sum_{i=0}^m N_i \dot{q}_i \quad (2.28c)$$

During the evolution of the system, the volume is conserved, which places a constraint on possible values of the $m + 1$ generalized coordinates. The system has only m independent generalized coordinates, which we choose as $\{q_1, \dots, q_m\}$. The condition of volume conservation can be expressed as

$$\delta V = \iint_{A_{14}} \delta r_n dA = \iint_{A_{14}} \sum_{i=0}^m N_i \delta q_i dA = \sum_{i=0}^m \delta q_i \iint_{A_{14}} \sum_{i=0}^m N_i dA = 0 \quad (2.29)$$

From which we obtain the virtual variation and actual velocity of the dependent generalized coordinate, δq_0 and $\dot{q}_0$, as

$$\delta q_0 = \frac{1}{\iint_{A_{14}} N_0 dA} \left(-\delta q_1 \iint_{A_{14}} N_1 dA - \dots - \delta q_m \iint_{A_{14}} N_m dA \right) = -\sum_{i=1}^m a_i \delta q_i \quad (2.30a)$$

$$\dot{q}_0 = \frac{1}{\iint_{A_{14}} N_0 dA} \left(-\dot{q}_1 \iint_{A_{14}} N_1 dA - \dots - \dot{q}_m \iint_{A_{14}} N_m dA \right) = -\sum_{i=1}^m a_i \dot{q}_i \quad (2.30b)$$

where the coefficient a_i is

$$a_i = \frac{\iint_{A_{14}} N_i dA}{\iint_{A_{14}} N_0 dA} \quad (i = 1, \dots, m) \quad (2.31)$$

Therefore, in term of the m independent generalized coordinates, the virtual motions and the actual velocities are expressed as

$$\delta r_n^{(t1)} = \sum_{i=0}^m N_i^{(t1)} \delta q_i = N_0^{(t1)} \delta q_0 + \sum_{i=1}^m N_i^{(t1)} \delta q_i = \sum_{i=1}^m (N_i^{(t1)} - N_0^{(t1)} a_i) \delta q_i = \sum_{i=1}^m \Phi_i^{(t1)} \delta q_i \quad (2.32a)$$

$$\delta r_n^{(t2)} = \sum_{i=0}^m N_i^{(t2)} \delta q_i = N_0^{(t2)} \delta q_0 + \sum_{i=1}^m N_i^{(t2)} \delta q_i = \sum_{i=1}^m (N_i^{(t2)} - N_0^{(t2)} a_i) \delta q_i = \sum_{i=1}^m \Phi_i^{(t2)} \delta q_i \quad (2.32b)$$

$$\delta r_n = \sum_{i=0}^m N_i \delta q_i = N_0 \delta q_0 + \sum_{i=1}^m N_i \delta q_i = \sum_{i=1}^m (N_i - N_0 a_i) \delta q_i = \sum_{i=1}^m \Phi_i \delta q_i \quad (2.32c)$$

$$v_n^{(t1)} = \sum_{i=0}^m N_i^{(t1)} \dot{q}_i = \sum_{i=1}^m \Phi_i^{(t1)} \dot{q}_i \quad (2.33a)$$

$$v_n^{(t2)} = \sum_{i=0}^m N_i^{(t2)} \dot{q}_i = \sum_{i=1}^m \Phi_i^{(t2)} \dot{q}_i \quad (2.33b)$$

$$v_n = \sum_{i=0}^m N_i \dot{q}_i = \sum_{i=1}^m \Phi_i \dot{q}_i \quad (2.33c)$$

where $\Phi_i^{(r1)}$, $\Phi_i^{(r2)}$, and Φ_i ($i = 1, \dots, m$) are the shape functions corresponding to the m independent generalized coordinate $\{q_1, \dots, q_m\}$, which also depend on the independent generalized coordinates $\{q_1, \dots, q_m\}$ but not on the generalized velocities.

The free energy is a function of the generalized coordinates, $G = G(q_0, q_1, \dots, q_m)$. By applying the constraint condition of volume conservation, we get

$$\delta G = \sum_{i=0}^m \frac{\partial G}{\partial q_i} \delta q_i = \frac{\partial G}{\partial q_0} \delta q_0 + \sum_{i=0}^m \frac{\partial G}{\partial q_i} \delta q_i = \sum_{i=1}^m -T_i \delta q_i \quad (2.34)$$

By comparing Eqs. (2.34) with the results from substituting (2.32) into Eq. (2.20), the generalized forces T_i ($i = 1, \dots, m$) are given by

$$T_i = \int_{L_{r1}} f_{t1} \Phi_i^{(r1)} ds_\psi + \int_{L_{r2}} f_{t2} \Phi_i^{(r2)} ds_l + \iint_{A_{r4}} f \Phi_i dA \quad (i = 1, \dots, m) \quad (2.35)$$

Substituting Eqs. (2.32), (2.33) and (2.34) into the weak statement, Eq. (2.26), gives

$$\sum_{i,j=1}^m H_{ij} \dot{q}_j \delta q_i = \sum_{i=1}^m T_i \delta q_i \quad (2.36)$$

where

$$H_{ij} = \int_{L_{r1}} \frac{\Phi_i^{(r1)} \Phi_j^{(r1)}}{M_{r1}} ds_\psi + \int_{L_{r2}} \frac{\Phi_i^{(r2)} \Phi_j^{(r2)}}{M_{r2}} ds_l + \iint_{A_{r4}} \frac{\Phi_i \Phi_j}{M} dA \quad (i, j = 1, \dots, m) \quad (2.37)$$

Eq. (2.36) holds true for arbitrary virtual changes δq_i , so that the coefficient for each δq_i must equal. Thus

$$\sum_{j=1}^m H_{ij} \dot{q}_j = T_i \quad (i = 1, \dots, m) \quad (2.38)$$

From Eqs. (2.35) and (2.37), it can be seen that H_{ij} and T_i are independent of the generalized velocities $\dot{q}_i$, so that Eq. (2.38) is a set of linear algebra equations for the

generalized velocities $\{\dot{q}_1, \dots, \dot{q}_m\}$. Once solved, $\dot{q}_0$ can be calculated according to Eq. (2.30b). Then, $\{\dot{q}_0, \dot{q}_1, \dots, \dot{q}_m\}$ update the generalized coordinates $\{q_0, q_1, \dots, q_m\}$ for a small time step. The process is repeated for many steps to evolve the system. Because H_{ij} and T_i depend on the generalized coordinates q_i , Eq. (2.38) is a set of first-order nonlinear ODEs with the variations of the generalized coordinates q_i to describe the evolution process of the system driven by surface tensions.

The physical significance of H_{ij} as a viscosity matrix now becomes evident from Eq. (2.38): H_{ij} is the resistance force in the q_i direction when the system state moves at unit generalized velocity in q_j direction. Because the LHS of Eq. (2.36), $\sum_{i,j=1}^m H_{ij} \dot{q}_j \delta q_i$, is the dissipation term related to virtual motion δq_i , and must be positive as required by the Second Law of Thermodynamics. Therefore, the viscosity matrix is symmetric and positive-definite.

At a point on the energy landscape $G(q_1, \dots, q_m)$ (the energy landscape is a surface in a hyperspace with the free energy as the vertical axis, and the independent generalized coordinates $q_1, \dots, q_m$ as the horizontal axes), the generalized force vector points to the direction of the steepest decent. According to Eq. (2.38), the generalized velocity vector points in a different direction, unless the viscosity matrix happens to be isotropic (i.e., a diagonal matrix with identical elements). Consequently, the evolution path does not usually follow the trajectory of the steepest decent. Since H_{ij} is positive-definite,

$$-\dot{G} = \sum_{i=1}^m \dot{q}_i T_i = \sum_{i,j=1}^m \dot{q}_i H_{ij} \dot{q}_j > 0 \quad (2.39)$$

Consequently, for a quasi-static non-equilibrium thermodynamic evolution process under constant temperature and constant external loading, the total free energy is strictly decreasing. One geometrical explanation of Eq. (2.39) is: the combination of the Second Law of Thermodynamics and linear kinetics requires that the generalized velocity and forces vectors make an acute angle.

2.5 Numerical simulation

By applying the model outlined in above sections, we simulate numerically the evolution process of a liquid bridge distributing periodically along the cylinder axis between a rigid cylinder and a rigid half space. The system evolves the shape of the liquid to the equilibrium state driven by the specific surface tensions.

The Galerkin approach outlined above suggests a global characterization, i.e., the computations are robust and insensitive to the local details of material structures [36, 39, 40]. One may describe the microstructures at any desired level of approximation, and any kind of generalized coordinates can be chosen, so long as they reasonably describe the overall shape. It is unnecessary to exactly describe the shape at “selective” locations, such as triple junctions. Therefore, at a given time, we assume that the meridian meniscus profile is still an arc of circle, and the two angles that this arc of circle make with the cylinder and the half space are equal to the two equilibrium contact angle θ_1 and θ_2 , respectively. Let us assume the filling angle to be described by the following parametric equation at a given time:

$$\psi = \psi_0 \sum_{k=0}^m \left(q_k \cos \frac{2k\pi y}{\lambda} \right) \quad (2.40)$$

where ψ_0 is the average filling angle at initial state, λ the wavelength, q_k ($k = 0, 1, \dots, m$) the generalized coordinates (including a dependent generalized coordinate that depends m independent generalized coordinates), which are functions of time t . It is noted that we only retain $m + 1$ terms in the parametric equation, so Eq. (2.40) only describes an approximate distribution of the filling angle. If one takes m sufficiently large, ψ can be as close to the accurate distribution as possible.

It is noted that the liquid bridge distributes periodically along the y -axis, and the liquid in each wavelength is centrally symmetric. Therefore, we only need to consider the liquid in the domain $y \in [0, \lambda / 2]$.

In the plane $y = \text{const.}$, the radius of the meridian meniscus profile R_1 and the half width of the liquid bridge area l are still given by Eqs. (2.3) and (2.5), but it vary with y . For the variation $\delta\psi$ of filling angle, we have

$$\delta l = \frac{\partial l}{\partial \psi} \delta \psi \quad (2.41)$$

In the plane $y = \text{const.}$, the curvature center (x_0, z_0) of the meridian meniscus profile is given by

$$x_0 = l + R_1 \sin \theta_2 \quad (2.42a)$$

$$z_0 = R_1 \cos \theta_2 \quad (2.42b)$$

The equation describing the meniscus is

$$x = x(\psi(y), z) = x(y, z) = x_0 - \sqrt{R_1^2 - (z_0 - z)^2} \quad (2.43)$$

As the filling angle ψ is small, we can expand the Eq. (2.43) as a series of ψ and differentiate the series with respect to y , giving

$$\frac{\partial x}{\partial y} = \frac{\partial x}{\partial \psi} \frac{\partial \psi}{\partial y} = (R + O(\psi)) \frac{\partial \psi}{\partial y} \approx R \frac{\partial \psi}{\partial y} \quad (2.44)$$

where

$$\frac{\partial \psi}{\partial y} = -\psi_0 \sum_{k=1}^m q_k \frac{2k\pi}{\lambda} \sin \frac{2k\pi y}{\lambda} \quad (2.45)$$

By geometrical analysis (see Fig. 2.2), the intersection area of the liquid in the plane $y = \text{const.}$ is given as

$$\begin{aligned} S_y = & \frac{1}{2} R^2 \sin \psi \cos \psi + \frac{1}{2} (R \sin \psi + l) R (1 - \cos \psi) - \frac{1}{2} R_1^2 (\pi - \psi - \theta_1 - \theta_2) \\ & + \frac{1}{2} R_1^2 \sin(\pi - \psi - \theta_1 - \theta_2) - \frac{1}{2} R^2 \psi \end{aligned} \quad (2.46)$$

Its variation with $\delta\psi$ is

$$\delta S_y = \frac{\partial S_y}{\partial \psi} \delta\psi \quad (2.47)$$

In the domain $[0, \lambda/2]$, the volume variation of the liquid with $\delta\psi$ is

$$\delta V = \iint_{A_{14}} \delta r_n dA = \int_0^{\lambda/2} \delta(S_y) dy = \int_0^{\lambda/2} \frac{\partial(S_y)}{\partial \psi} \delta\psi dy = \int_0^{\lambda/2} \frac{\partial(S_y)}{\partial \psi} \sum_{j=0}^m \frac{\partial \psi}{\partial q_j} \delta q_j dy \quad (2.48)$$

where

$$\frac{\partial \psi}{\partial q_j} = \psi_0 \cos \frac{2j\pi y}{\lambda} \quad (j = 0, 1, \dots, m) \quad (2.49)$$

From Eq. (2.48) and the constraint condition of volume conservation, δq_0 is expressed as a function of m independent generalized coordinates as

$$\delta q_0 = - \frac{1}{\int_0^{\lambda/2} \frac{\partial(S_y)}{\partial \psi} dy} \sum_{i=1}^m \left(\delta q_i \int_0^{\lambda/2} \frac{\partial(S_y)}{\partial \psi} \cos \frac{2i\pi y}{\lambda} dy \right) = - \sum_{i=1}^m (a_i \delta q_i) \quad (2.50)$$

where

$$a_i = \frac{1}{\int_0^{\lambda/2} \frac{\partial(S_y)}{\partial \psi} dy} \int_0^{\lambda/2} \frac{\partial(S_y)}{\partial \psi} \cos \frac{2i\pi y}{\lambda} dy \quad (i = 0, 1, \dots, m) \quad (2.51)$$

At L_{t1} , the partial derivatives of $x = f(y, z)$ describing the meniscus with respect to y and z are

$$x_{,y} \approx R \frac{\partial \psi}{\partial y} \quad (2.52a)$$

$$x_{,z} = \text{ctg}(\theta_1 + \psi) \quad (2.52b)$$

Substituting Eq. (2.52) into Eq. (2.16) gives the current contact angle between the meniscus and the cylinder surface as

$$\cos(\theta_1(y)) = (\sin \psi + \text{ctg}(\theta_1 + \psi) \cos \psi) / \sqrt{1 + R^2 \left(\frac{\partial \psi}{\partial y} \right)^2 + \text{ctg}^2(\theta_1 + \psi)} \quad (2.53)$$

Similarly, at L_{12} , the partial derivatives of $x = f(y, z)$ with respect to y and z are

$$x_{,y} = l_{,y} = \frac{\partial l}{\partial \psi} \frac{\partial \psi}{\partial y} \quad (2.54a)$$

$$x_{,z} = -\text{ctg} \theta_2 \quad (2.54b)$$

Substituting Eq. (2.54) into Eq. (2.17) gives the current contact angle between the meniscus and the half space surface as

$$\cos(\theta_2(y)) = \text{ctg} \theta_2 / \sqrt{1 + \left(\frac{\partial l}{\partial \psi} \right)^2 \left(\frac{\partial \psi}{\partial y} \right)^2 + \text{ctg}^2 \theta_2} \quad (2.55)$$

The accurate value of the mean curvature of the meniscus κ can be calculated from Eqs. (2.11) and (2.12), but in this numerical simulation, we only calculate κ approximately as follows.

We assume that κ is constant on the meniscus profile with $y = \text{const.}$. As the filling angle ψ is small, we can ignore the contribution of the second principal curvature radius, R_2 , to κ . It is noticed that the height of the liquid bridge is $R(1 - \cos \psi)$, so κ is given approximately by

$$\kappa = \frac{\cos(\theta_1(y) + \psi_y) + \cos(\theta_2(y))}{R(1 - \cos \psi)} = \frac{\cos(\theta_1(y))\cos(\psi_y) - \sin(\theta_1(y))\sin(\psi_y) + \cos(\theta_2(y))}{R(1 - \cos \psi)} \quad (2.56)$$

where ψ_y is calculated by

$$\sin(\psi_y) = \frac{\sin \psi}{\sqrt{1 + \left(R \frac{\partial \psi}{\partial y}\right)^2}} \quad (2.57a)$$

$$\cos(\psi_y) = \sqrt{1 - \sin^2(\psi_y)} \quad (2.57b)$$

as the meniscus profile varies along y . $\sin(\theta_1(y))$ in Eq. (2.56) is given as

$$\sin(\theta_1(y)) = \sqrt{1 - (\cos(\theta_1(y)))^2} \quad (2.58)$$

The driving forces for the motions of the meniscus and two TPLs, f , f_{t1} , and f_{t2} , can be calculate from Eqs. (2.22).

For the liquid bridge in the domain $[y, y + dy]$, we consider the volume variation due to the variations of the generalized coordinates, giving

$$\begin{aligned} \iint_{A_{14}} \delta r_n dA &= \int_y^{y+dy} \delta(S_y) dy = \int_y^{y+dy} \frac{\partial S_y}{\partial \psi} \sum_{i=0}^m \left(\frac{\partial \psi}{\partial q_i} \delta q_i \right) dy \\ &= \int_y^{y+dy} \frac{\partial S_y}{\partial \psi} \sum_{i=1}^m \delta q_i \left(\frac{\partial \psi}{\partial q_i} - \frac{\partial \psi}{\partial q_0} a_i \right) dy \\ &= \int_y^{y+dy} \sum_{i=1}^m M_i \delta q_i dy \end{aligned} \quad (2.59)$$

If we assume that the actual velocity of the meniscus v_n is constant in the plane $y = \text{const.}$, we obtain

$$\iint_{A_{14}} v_n dA \approx v_n R_1 (\pi - \theta_1 - \theta_2 - \psi) \sqrt{1 + \left(R \frac{\partial \psi}{\partial y}\right)^2} dy = \int_y^{y+dy} \frac{dS_y}{dt} dy = \frac{dS_y}{dt} dy = \frac{\partial S_y}{\partial \psi} \sum_{i=0}^m \left(\frac{\partial \psi}{\partial q_i} \dot{q}_i \right) dy \quad (2.60)$$

Therefore, the approximate value of v_n in the plane $y = \text{const.}$ is

$$\begin{aligned}
v_n &\approx \frac{\frac{\partial S_y}{\partial \psi} \sum_{i=0}^m \left(\frac{\partial \psi}{\partial q_i} \dot{q}_i \right)}{R_1(\pi - \theta_1 - \theta_2 - \psi) \sqrt{1 + \left(R \frac{\partial \psi}{\partial y} \right)^2}} \\
&= \frac{\frac{\partial S_y}{\partial \psi}}{R_1(\pi - \theta_1 - \theta_2 - \psi) \sqrt{1 + \left(R \frac{\partial \psi}{\partial y} \right)^2}} \sum_{i=1}^m \left(\frac{\partial \psi}{\partial q_i} - \frac{\partial \psi}{\partial q_0} a_i \right) \dot{q}_i \\
&= \sum_{i=1}^m \Phi_i \dot{q}_i
\end{aligned} \tag{2.61}$$

The vector equation describing L_{t1} is

$$\bar{x}^{(t1)} = R \sin \psi \bar{i} + y \bar{j} + R(1 - \cos \psi) \bar{k} \tag{2.62}$$

Its tangential vector is

$$\frac{\partial \bar{x}^{(t1)}}{\partial y} = R \cos \psi \frac{\partial \psi}{\partial y} \bar{i} + \bar{j} + R \sin \psi \frac{\partial \psi}{\partial y} \bar{k} \tag{2.63}$$

As the unit vector normal to the cylinder surface directing to the vapor phase is $\sin \psi \bar{i} - \cos \psi \bar{k}$, the unit vector normal to L_{t1} in the cylinder surface directing from the liquid phase to the vapor phase is given as

$$\bar{n}^{(t1)} = \frac{(\sin \psi \bar{i} - \cos \psi \bar{k}) \times \frac{\partial \bar{x}^{(t1)}}{\partial y}}{\left| \frac{\partial \bar{x}^{(t1)}}{\partial y} \right|} = \frac{\sin \psi \bar{k} - R \psi_{,y} \bar{j} + \cos \psi \bar{i}}{\sqrt{1 + R^2 (\psi_{,y})^2}} \tag{2.64}$$

Therefore, from Eq. (2.27a), we obtain

$$\begin{aligned}
\delta r_n^{(t1)} &= \sum_{i=0}^m \left(\bar{n}^{(t1)} \cdot \frac{\partial \bar{x}^{(t1)}}{\partial q_i} \right) \delta q_i \equiv \sum_{i=0}^m R \frac{\partial \psi}{\partial q_i} / \sqrt{1 + \left(R \frac{\partial \psi}{\partial y} \right)^2} \delta q_i \\
&= \sum_{i=1}^m R \left(\frac{\partial \psi}{\partial q_i} - \frac{\partial \psi}{\partial q_0} a_i \right) \delta q_i / \sqrt{1 + \left(R \frac{\partial \psi}{\partial y} \right)^2} = \sum_{i=1}^m \Phi_i^{(t1)} \delta q_i
\end{aligned} \tag{2.65}$$

The vector equation describing L_{r2} is

$$\bar{x}^{(r2)} = l\bar{i} + y\bar{j} \quad (2.66)$$

The unit vector normal to L_{r2} in the half space surface directing from the liquid phase to the vapor phase is

$$\bar{n}^{(r2)} = (\bar{i} - l_y \bar{j}) / \sqrt{1 + (l_y)^2} \quad (2.67)$$

where l, y is given by Eq. (2.54a).

Therefore, from Eq. (2.27b), we obtain

$$\begin{aligned} \delta r_n^{(r2)} &= \sum_{i=0}^m \left(\bar{n}^{(r2)} \cdot \frac{\partial \bar{x}^{(r2)}}{\partial q_i} \right) \delta q_i = \sum_{i=0}^m \frac{\partial l}{\partial \psi} \frac{\partial \psi}{\partial q_i} \delta q_i / \sqrt{1 + (l_y)^2} \\ &= \frac{\partial l}{\partial \psi} \sum_{i=1}^m \left(\frac{\partial \psi}{\partial q_i} - \frac{\partial \psi}{\partial q_0} a_i \right) \delta q_i / \sqrt{1 + (l_y)^2} = \sum_{i=1}^m \Phi_i^{(r2)} \delta q_i \end{aligned} \quad (2.68)$$

From Eqs. (2.65) and (2.68), the relations between the actual velocities and the independent generalized velocities at two TPLs are given as

$$v_n^{(r1)} = \sum_{i=1}^m \Phi_i^{(r1)} \dot{q}_i, \quad v_n^{(r2)} = \sum_{i=1}^m \Phi_i^{(r2)} \dot{q}_i \quad (2.69)$$

Substituting $\delta r_n^{(r1)}$ (Eq. (2.65)), $\delta r_n^{(r2)}$ (Eq. (2.68)) and δr_n (Eq. (2.59)) into Eq. (2.20) gives the free energy variation due to $\delta r_n^{(r1)}$, $\delta r_n^{(r2)}$ and δr_n as

$$\delta G = - \int_{L_{r1}} f_{r1} \sum_{i=1}^m \Phi_i^{(r1)} \delta q_i ds_\psi - \int_{L_{r2}} f_{r2} \sum_{i=1}^m \Phi_i^{(r2)} \delta q_i ds_l - \int_0^{\lambda/2} f \sum_{i=1}^m M_i \delta q_i dy \quad (2.70)$$

Substitute $\delta r_n^{(r1)}$ (Eq. (2.65)), $\delta r_n^{(r2)}$ (Eq. (2.68)), δr_n (Eq. (2.59)), $v_n^{(r1)}$ and $v_n^{(r2)}$ (Eqs. (2.69)), and v_n (Eq. (2.61)) into the LHS of Eq. (2.26) for the weak statement, and substitute Eq. (2.70) into the RHS of Eq. (2.26), giving

$$\sum_{i,j=1}^m H_{ij} \dot{q}_j \delta q_i = \sum_{i=1}^m T_i \delta q_i \quad (2.71)$$

with

$$H_{ij} = \int_{L_{i1}} \frac{\Phi_i^{(r1)} \Phi_j^{(r1)}}{M_{i1}} ds_\psi + \int_{L_{i2}} \frac{\Phi_i^{(r2)} \Phi_j^{(r2)}}{M_{i2}} ds_l + \int_0^{\lambda/2} \frac{M_i \Phi_j}{M} dy \quad (i, j = 1, \dots, m) \quad (2.72)$$

$$T_i = \int_{L_{i1}} f_{i1} \Phi_i^{(r1)} ds_\psi + \int_{L_{i2}} f_{i2} \Phi_i^{(r2)} ds_l + \int_0^{\lambda/2} f M_i dy \quad (i = 1, \dots, m) \quad (2.73)$$

Eq. (2.71) must hold true for any virtual motion δq_i ($i = 1, \dots, m$), so that

$$\sum_{j=1}^m H_{ij} \dot{q}_j = T_i \quad (i = 1, \dots, m) \quad (2.74)$$

From Eqs. (2.73) and (2.74), it is noticed that H_{ij} and T_i only depend on the generalized coordinate $\{q_1, \dots, q_m\}$ nonlinearly but not on the generalized velocities. Eq. (2.74) is the governing equations of the evolution process. By solving this set of ODEs under initial conditions of the generalized coordinates, the history of the generalized coordinates with respect to time t describes the entire evolution process.

All the terms in Eq. (2.73) and (2.74) are listed as follows:

$$\int_{L_{i1}} \frac{\Phi_i^{(r1)} \Phi_j^{(r1)}}{M_{i1}} ds_\psi = \frac{R^2 \psi_0^2}{M_{i1}} \int_0^{\lambda/2} \frac{\left(\cos \frac{2i\pi y}{\lambda} - a_i \right)}{\sqrt{1 + \left(R \frac{\partial \psi}{\partial y} \right)^2}} \left(\cos \frac{2j\pi y}{\lambda} - a_j \right) dy \quad (2.75a)$$

$$\int_{L_{i2}} \frac{\Phi_i^{(r2)} \Phi_j^{(r2)}}{M_{i2}} ds_l = \frac{\psi_0^2}{M_{i2}} \int_0^{\lambda/2} \frac{\left(\frac{\partial l}{\partial \psi} \right)^2 \left(\cos \frac{2i\pi y}{\lambda} - a_i \right)}{\sqrt{1 + \left(\frac{\partial l}{\partial \psi} \right)^2 \left(\frac{\partial \psi}{\partial y} \right)^2}} \left(\cos \frac{2j\pi y}{\lambda} - a_j \right) dy \quad (2.75b)$$

$$\int_0^{\lambda/2} \frac{M_i \Phi_j}{M} dy = \frac{\psi_0^2}{M} \int_0^{\lambda/2} \frac{\left(\frac{\partial S_y}{\partial \psi} \right)^2 \left(\cos \frac{2i\pi y}{\lambda} - a_i \right)}{R_1 (\pi - \theta_1 - \theta_2 - \psi) \sqrt{1 + \left(R \frac{\partial \psi}{\partial y} \right)^2}} \left(\cos \frac{2j\pi y}{\lambda} - a_j \right) dy \quad (2.75c)$$

$$\begin{aligned}
\int_{L_{t1}} f_{t1} \Phi_i^{(t1)} ds_\psi &= R\psi_0 \int_0^{\lambda/2} f_{t1} \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) dy \\
&= R\gamma_{14}\psi_0 \int_0^{\lambda/2} (\cos \theta_1 - \cos \theta_1(y)) \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) dy
\end{aligned} \tag{2.75d}$$

$$\begin{aligned}
\int_{L_{t2}} f_{t2} \Phi_i^{(t2)} ds_l &= \psi_0 \int_0^{\lambda/2} f_{t2} \frac{\partial l}{\partial \psi} \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) dy \\
&= \gamma_{14}\psi_0 \int_0^{\lambda/2} (\cos \theta_2 - \cos \theta_2(y)) \frac{\partial l}{\partial \psi} \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) dy
\end{aligned} \tag{2.75e}$$

$$\begin{aligned}
\int_0^{\lambda/2} f M_i dy &= \psi_0 \int_0^{\lambda/2} f \left(\frac{\partial S_y}{\partial \psi} \right) \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) dy \\
&= \psi_0 \int_0^{\lambda/2} (\kappa\gamma_{14} - \Delta p) \left(\frac{\partial S_y}{\partial \psi} \right) \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) dy \\
&= \gamma_{14}\psi_0 \int_0^{\lambda/2} \kappa \left(\frac{\partial S_y}{\partial \psi} \right) \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) dy
\end{aligned} \tag{2.75f}$$

In order to study the evolution problem parametrically, Eq. (2.74) will be written in a dimensionless form. This evolution problem has seven dimensionless independent quantities, namely ψ_0 , θ_1 , θ_2 , $\tilde{M}_{t2}$, $\tilde{M}$, $\tilde{\lambda}$, and $\tilde{t}$.

$$\tilde{M}_{t2} = \frac{M_{t2}}{M_{t1}} \tag{2.76a}$$

$$\tilde{M} = \frac{M}{R\psi_0^2 M_{t1}} \tag{2.76b}$$

$$\tilde{\lambda} = \frac{\lambda}{R\psi_0} \tag{2.76c}$$

$$\tilde{t} = t / \left(\frac{R\psi_0}{M_{t1}\gamma_{14}} \right) = t / t_0 \tag{2.76d}$$

where we introduce a characteristic time t_0 as

$$t_0 = \frac{R\psi_0}{M_{t1}\gamma_{14}} \quad (2.77)$$

$\tilde{M}$ describes the ratio of the mobility of the meniscus to the mobilities of the two TPLs, and $\tilde{\lambda}$ describes the ratio of characterization length (λ) to the characterization width ($R\psi_0$) for the liquid bridge area. When $\tilde{M}$ is large, the evolution process is mainly controlled by the motions of two TPLs, as the meniscus moves more quickly than the TPLs. Conversely, when $\tilde{M}$ is small, the evolution process is mainly controlled by the motion of the meniscus, as the TPLs move more quickly than the meniscus.

With these dimensionless independent quantities, Eq. (2.74) can be rewritten in a dimensionless form:

$$\sum_{j=1}^m \tilde{H}_{ij} \frac{dq_j}{d\tilde{t}} = \tilde{T}_i \quad (i = 1, \dots, m) \quad (2.78)$$

with

$$\tilde{H}_{ij} = \int_0^{\lambda/2} \left[\frac{\left(\frac{\partial \left(\frac{l}{R\psi_0} \right)}{\partial \left(\frac{\psi}{\psi_0} \right)} \right)^2}{\sqrt{1 + \frac{1}{\tilde{\lambda}} \frac{\partial \left(\frac{\psi}{\psi_0} \right)}{\partial \left(\frac{y}{\lambda} \right)}}} + \frac{1}{\tilde{M}_{t2}} \frac{\left(\frac{\partial \left(\frac{l}{R\psi_0} \right)}{\partial \left(\frac{\psi}{\psi_0} \right)} \right)^2}{\sqrt{1 + \frac{1}{\tilde{\lambda}} \frac{\partial \left(\frac{l}{R\psi_0} \right)}{\partial \left(\frac{\psi}{\psi_0} \right)} \frac{\partial \left(\frac{\psi}{\psi_0} \right)}{\partial \left(\frac{y}{\lambda} \right)}}} + \frac{\left(\frac{\partial \left(\frac{S_y}{R^2\psi_0^3} \right)}{\partial \left(\frac{\psi}{\psi_0} \right)} \right)^2}{\sqrt{1 + \frac{1}{\tilde{\lambda}} \frac{\partial \left(\frac{\psi}{\psi_0} \right)}{\partial \left(\frac{y}{\lambda} \right)}}} + \frac{1}{\tilde{M}} \frac{\left(\frac{R_1}{R\psi_0^2} \right) \left(\pi - \theta_1 - \theta_2 - \left(\frac{\psi}{\psi_0} \right) \psi_0 \right)}{\sqrt{1 + \frac{1}{\tilde{\lambda}} \frac{\partial \left(\frac{\psi}{\psi_0} \right)}{\partial \left(\frac{y}{\lambda} \right)}}} \right] \left[\begin{array}{l} \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) \\ \times \left(\cos \frac{2j\pi y}{\lambda} - a_j \right) \end{array} \right] \frac{1}{\lambda} dy \quad (2.79)$$

$$\tilde{T}_i = \int_0^{\lambda/2} \left[\begin{aligned} & (\cos \theta_1 - \cos \theta_1(y)) + (\cos \theta_2 - \cos \theta_2(y)) \frac{\partial [l/(R\psi_0)]}{\partial [\psi/\psi_0]} \\ & + (\kappa R \psi_0^2) \left(\frac{\partial [S_y/(R^2 \psi_0^3)]}{\partial [\psi/\psi_0]} \right) \end{aligned} \right] \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) \frac{1}{\lambda} dy \quad (2.80)$$

According to the terminology of the non-equilibrium thermodynamics for micro-structural evolution [36, 39], the six dimensionless independent quantities $(\psi_0, \theta_1, \theta_2, \tilde{M}_{i2}, \tilde{M}, \text{ and } \tilde{\lambda})$ are the control parameters, which control the structural evolution in the material system. Once they are given, we can solve Eq. (2.78) under certain initial conditions for $\{q_0, q_1, \dots, q_m\}$ to obtain the history of the generalized coordinates $\{q_1, \dots, q_m\}$.

To solve Eq. (2.78), the Simpson method can be used to evaluate the integrals in Eqs. (2.79) and (2.80), and the fourth-order Runge-Kutta method can be used to solve the set of ODEs [Eq. (2.78)] for $q_i(t)$ ($i = 1, \dots, m$), then Eq. (2.50) is used to obtain $q_0(t)$ after each time step.

In the following, we discuss two extreme cases: the dimensionless quantity $\tilde{M}$ is infinite and infinitesimal.

When $\tilde{M}$ is infinite, Setting Eq. (2.75c) as zero, Eq. (2.79) is simplified as

$$\tilde{H}_{ij} = \int_0^{\lambda/2} \left[\frac{1}{\sqrt{1 + \left(\frac{1}{\tilde{\lambda}} \frac{\partial \left(\frac{\psi}{\psi_0} \right)}{\partial \left(\frac{y}{\lambda} \right)} \right)^2}} + \frac{1}{\tilde{M}_{i2}} \frac{\left(\frac{\partial \left(\frac{l}{R\psi_0} \right)}{\partial \left(\frac{\psi}{\psi_0} \right)} \right)^2}{\sqrt{1 + \left(\frac{1}{\tilde{\lambda}} \frac{\partial \left(\frac{l}{R\psi_0} \right) \frac{\partial \left(\frac{\psi}{\psi_0} \right)}{\partial \left(\frac{y}{\lambda} \right)} \right)^2}} \right] \left[\begin{aligned} & \left(\cos \frac{2i\pi y}{\lambda} - a_i \right) \\ & \times \left(\cos \frac{2j\pi y}{\lambda} - a_j \right) \end{aligned} \right] \frac{1}{\lambda} dy \quad (2.81)$$

Eq. (2.78) and Eq. (2.80) are still applicable.

Conversely, when $\tilde{M}$ is infinitesimal, Eq. (2.75a) and Eq. (2.75b) are equal to zero. If the dimensionless time is changed as

$$\tilde{t} = t / \left(\frac{R^2 \psi_0^3}{M \gamma_{14}} \right) \quad (2.82)$$

and the dimensionless independent quantities $\psi_0, \theta_1, \theta_2, \tilde{M}_{i2} = \frac{M_{i2}}{M_{i1}}, \tilde{\lambda} = \frac{\lambda}{R \psi_0}$ are

still adopted, the dimensionless viscosity matrix $\tilde{H}_{ij}$ in Eq. (2.78) is given by

$$\tilde{H}_{ij} = \int_0^{\lambda/2} \left[\frac{\left(\frac{\partial \left(\frac{S_y}{R^2 \psi_0^3} \right)}{\partial \left(\frac{\psi}{\psi_0} \right)} \right)^2}{\left(\frac{R_1}{R \psi_0^2} \right) \left(\pi - \theta_1 - \theta_2 - \left(\frac{\psi}{\psi_0} \right) \psi_0 \right) \sqrt{1 + \left(\frac{1}{\tilde{\lambda}} \frac{\partial \left(\frac{\psi}{\psi_0} \right)}{\partial \left(\frac{y}{\lambda} \right)} \right)^2}} \right] \left[\left(\cos \frac{2i\pi y}{\lambda} - a_i \right) \times \left(\cos \frac{2j\pi y}{\lambda} - a_j \right) \right] \frac{1}{\lambda} dy \quad (2.83)$$

The expression for $\tilde{T}_i$ is still Eq. (2.80).

2.6 Numerical results and analysis

We have performed many case studies to check our program and to analyze the effects of control parameters on the evolution of the liquid bridge between a rigid cylinder and a rigid plane. As the mobilities of the two TPLs are determined by similar mechanism, without losing generality, we assumed that the cylinder and the plane are made of identical materials, i.e., $M_t = M_{t1} = M_{t2}$ and $\theta = \theta_1 = \theta_2$, so that we can discuss the competition between the mobilities of the meniscus and the TPLs in the evolution process of the liquid bridge. Therefore, the six dimensionless control parameters

reduce to four, i.e., ψ_0 , θ , $\tilde{M}$ and $\tilde{\lambda}$. The discussion on the effects of the control parameters on the evolution process can be simplified.

In all the case studies, we assume the filling angle at the beginning as

$$\psi = \psi_0 \left(1 + 0.99 \cos \frac{2\pi y}{\lambda} \right) \quad (2.84)$$

where ψ_0 is the average filling angle at initial state. A comparison between Eq. (2.84) and Eq. (2.40) gives the initial values of the generalized coordinates as

$$\begin{cases} q_0 = 1 & q_1 = 0.99 \\ q_i = 0 & \text{for } i = 2, \dots, m \end{cases} \quad (2.85)$$

where q_1 is assumed as 0.99 so that a continuous liquid bridge exists between the cylinder and the plane.

The variation of the liquid bridge volume is monitored in the simulations. It is less than 10^{-4} time of the initial volume for all the cases, and the liquid bridge volume can be taken to be conserved.

The numerical simulation results show that the evolution process is extremely rapid at its beginning in a very short time, then followed by a slow process in a long time before reaching its final equilibrium state. The maximum, average and minimum of the filling angle all reach the same value when the liquid bridge reach its final equilibrium state after a long evolution process.

The final equilibrium configurations of the liquid bridge are same as that obtained by solving Laplace-Young equation in Section 2.2. Therefore, by using our theoretical model, we can predict the liquid bridge configurations at both the final equilibrium state and any time during the evolution process.

As expected, the final equilibrium configurations of the liquid bridge are same for all the cases with same values of ψ_0 and θ , and is not affected by the values of $\tilde{M}$ and $\tilde{\lambda}$.

Fig. 2.4 shows the effects of the dimensionless wavelength $\tilde{\lambda}$ of the initial filling angle on the liquid bridge configuration (the minimum, average and maximum of the filling angle: $\psi_{\min}$, ψ_{aver} , $\psi_{\max}$) and the free energy of the system (G). The values of the filling angle are normalized by the equilibrium contact angle ψ_{eq} . The free energy of the system is calculated with reference to the final equilibrium state and is normalized by the free energy of the system G_0 without liquid bridge. For demonstration purpose, the initial average filling angle and contact angle are taken to be $\psi_0 = 2^\circ$ and $\theta = 60^\circ$; results for cases with different values of ψ_0 and θ are similar to these cases. $\tilde{M}$ is taken to be 0, 1, and ∞ . The effect of $\tilde{\lambda}$ on the evolution process is ignorable when $\tilde{\lambda} \geq 10$. The smaller the dimensionless wavelength of the liquid bridge, the more quickly the system evolves and the shorter the time needed for the system to reach the final equilibrium state. The effect of $\tilde{\lambda}$ increases with $\tilde{M}$ increasing from 0 to ∞ . The estimated values of the normalized equilibrium times needed for the system to reach its final equilibrium state, $\tilde{t}_{eq} = t_{eq}/t_0$, are listed in Table 2.1 for different values of $\tilde{\lambda}$. It should be noted that the characterization times t_0 are $t_0 = (R\psi_0)/(M\gamma_{14})$ for $\tilde{M} = 1$ and ∞ and $t_0 = (R^2\psi_0^3)/(M\gamma_{14})$ for $\tilde{M} = 0$.

Table 2.1 The normalized equilibrium times for different $\tilde{\lambda}$

	$\tilde{\lambda} = 1$	$\tilde{\lambda} = 10$	$\tilde{\lambda} = 100$	$\tilde{\lambda} = 1000$
$\tilde{M} = 0$	4.35035	4.63024	4.63024	4.63024
$\tilde{M} = 1$	14.73813	16.87915	16.87915	16.87915
$\tilde{M} = \infty$	9.86709	11.59912	11.78896	11.78896

Fig. 2.5 shows the effects of the dimensionless mobility $\tilde{M}$ on the liquid bridge configuration and the free energy of the system. The initial average filling angle and the contact angle are also taken to be $\psi_0 = 2^\circ$ and $\theta = 60^\circ$. As $\tilde{\lambda}$ has a small effect

on the evolution process when it has a small value, such as 1, we give the results for two different values of $\tilde{\lambda}$, i.e., $\tilde{\lambda}=1$ and 100. The latter shows the effect of $\tilde{M}$ when the effect of $\tilde{\lambda}$ can be ignored. For the convenience of discussion, we assume that the mobility of the TPL is fixed, i.e., $M_t = \text{const.}$. When the dimensionless mobility of the system $\tilde{M}$ decreases (i.e., the mobility of the meniscus M decreases), the evolution velocity decreases and the system needs more time to reach its final equilibrium state. The effect of $\tilde{M}$ becomes large especially for the cases with $\tilde{M}$ much smaller than 1. The estimated values of the normalized equilibrium times needed for the system to reach its final equilibrium state are listed in Table 2.2 for different values of $\tilde{M}$. All the equilibrium times t_{eq} are normalized by $t_0 = (R\psi_0)/(M_t\gamma_{14})$. Therefore, because we assume that $M_t = \text{const.}$, the equilibrium time increases up to infinite when $\tilde{M}$ reduces to zero.

Table 2.2 The normalized equilibrium times for different $\tilde{M}$

	$\tilde{M} = \infty$	$\tilde{M} = 100$	$\tilde{M} = 1$	$\tilde{M} = 0.01$	$\tilde{M} = 0$
$\tilde{\lambda} = 1$	9.86709	9.86709	14.73813	492.6281	∞
$\tilde{\lambda} = 100$	11.78896	11.78896	16.87915	545.063	∞

Fig. 2.6 shows the effects of the initial average filling angle on the evolution process. The contact angle is fixed, i.e., $\theta = 60^\circ$. The dimensionless wavelength $\tilde{\lambda}$ is taken to be 1 and 100 and the dimensionless mobility $\tilde{M}$ is taken to be $\tilde{M}=0, 1$ and infinite. The evolution processes are compared for two different filling angles $\psi_0=0.2^\circ$ and 2° . The initial average filling angle has small effect on the evolution process only when $\tilde{\lambda}=1$. When $\tilde{\lambda}$ is large as 100, its effect on the liquid bridge configuration and the free energy of the system is ignorable.

Fig. 2.7 shows the effects of the equilibrium contact angle θ on the evolution process. The initial average filling angle is taken to be $\theta = 2^\circ$. Smaller contact angles induce

larger driving forces on the TPLs and the meniscus, so that the evolution process proceeds more rapidly, and the time needed for the system to reach its final equilibrium state becomes shorter. The estimated values of the normalized equilibrium times $\tilde{t}_{eq} = t_{eq}/t_0$ for different contact angles are listed in Table 2.3. As mentioned above, the characterization times t_0 are $t_0 = (R\psi_0)/(M\gamma_{14})$ for $\tilde{M} = 1$ and ∞ and $t_0 = (R^2\psi_0^3)/(M\gamma_{14})$ for $\tilde{M} = 0$.

Table 2.3 The normalized equilibrium times for different θ

	$\theta = 10^\circ$	$\theta = 30^\circ$	$\theta = 60^\circ$	$\theta = 80^\circ$
$\tilde{M} = 0; \tilde{\lambda} = 1$	1.68098	2.34098	4.35035	14.08837
$\tilde{M} = 0; \tilde{\lambda} = 100$	1.85525	2.45176	4.63024	14.30372
$\tilde{M} = 1; \tilde{\lambda} = 1$	6.65098	8.46689	14.73813	43.30154
$\tilde{M} = 1; \tilde{\lambda} = 100$	7.69941	9.74814	16.87915	50.71732
$\tilde{M} = \infty; \tilde{\lambda} = 1$	5.74043	6.06452	9.86709	33.09596
$\tilde{M} = \infty; \tilde{\lambda} = 100$	6.71856	7.09824	11.78896	35.05966

2.7 Summary and discussion

In the present work, a theoretical model was set up to analyze the kinetic evolution process of a liquid bridge between a rigid cylinder and a rigid half space. By following the irreversible non-equilibrium thermodynamics framework for micro-structural evolution [36, 39], the time-dependent evolution of the liquid bridge driven by surface tensions was modeled by a nonlinear dynamic system. It leads to a set of nonlinear ODEs with generalized coordinates. Based on the initial conditions of the generalized coordinates, solutions to the ODEs describe the evolution history.

The reduction in the free energy of the system drives the thermodynamics evolution process. Virtual motions are small movements on the meniscus or the TPLs, which conserve the mass of the system. The thermodynamics forces are the free energy decrease associated with unit virtual motions. The actual velocities of the meniscus and TPLs are related to the thermodynamics force according to the kinetic laws. The free energy, motion descriptions, driving forces and kinetic laws define a nonlinear dynamic system for the thermodynamics evolution.

The nonlinear dynamic system can be formulated by a weak statement, which leads to the Galerkin method in numerical analysis. By describing the system with $m + 1$ generalized coordinates $\{q_0, q_1, \dots, q_m\}$, a global approach is used to obtain an approximate solutions to the time-dependent evolution problem.

The evolution process of the liquid bridge between a rigid cylinder and a rigid half space is controlled by 4 dimensionless control parameters, i.e., the initial average filling angle ψ_0 , the equilibrium contact angle θ , the dimensionless mobility $M / (R\psi_0^2 M_t)$, and the dimensionless wavelength of the liquid bridge.

The filling angle ψ_0 has a small effect on the evolution process of the non-equilibrium liquid bridge, especially when the dimensionless wavelength $\lambda / (R\psi_0)$ is larger than 10. The effect of the dimensionless wavelength $\lambda / (R\psi_0)$ on the evolution process is also small. Smaller the wavelength, more rapid the evolution process is. The evolution process is more rapid for a liquid bridge with a smaller contact angle θ . With the mobility of the TPLs being fixed, the non-equilibrium liquid bridge evolves at a slow velocity for small dimensionless mobility $\tilde{M}$.

In the present work, we have adopted a global approach to look at structural evolution. One may describe an evolving structure with any kind and any number of generalized coordinates, depending on one's purpose and the requirement of accuracy. A global approach uniformly treats all kinds of thermodynamics forces and rate processes; it readily handles surface tension anisotropy; it avoids the issue of local equilibrium, which comes out as a consequence. This approach reduces a continuum structure to a low-dimension dynamical system.

Our numerical results show that the liquid droplets will evolve to an equilibrium liquid bridge, which induces a liquid bridging force due to surface tension. In micro-scale with small external load, this liquid bridging force becomes one of the dominant factors. It is both theoretically and practically important to understand the liquid bridging phenomenon, especially with the recent interests of MEMS development and applications.

Fig. 2.1 A liquid-bridging induced contact problem

Fig. 2.2 Geometrical parameters in liquid-bridging induced contact

Fig. 2.3 The meniscus and two TPLs undergo virtual motions δr_n , $\delta r_n^{(r1)}$, and $\delta r_n^{(r2)}$

Fig. 2.4 The effect of the wavelength on the liquid bridge configuration and the free energy of the system

(a)

(b)

Fig. 2.5 The effect of the mobility on the liquid bridge configuration and the free energy of the system

(c)

(d)

(k)

(l)

Fig. 2.6 The effect of the filling angle on the liquid bridge configuration and the free energy of the system

(c)

(d)

(i)

(j)

Fig. 2.7 The effect of the contact angle on the liquid bridge configuration and the free energy of the system

Chapter 3

Elastic Analysis for Liquid-Bridging Induced Contact

3.1 Introduction

As mentioned in Chapter 2, with the presence of a thin liquid film or a droplet of liquid at the contact interface, liquid bridging phenomenon has been observed in a wide range of engineering applications. Adhesive micro-contact induced by liquid bridging force is the most important factor determining the properties and lives of products in modern MEMS devices. An initially non-equilibrium liquid bridge will evolve to an equilibrium liquid bridge. In this chapter, we will focus on the contact induced by equilibrium liquid bridging as illustrated in Fig. 3.1a.

The liquid bridging has been observed in a wide range of engineering applications. Consequently, investigators from various fields have studied various aspects of the liquid bridging phenomenon [5, 12, 14], where the solids were considered as rigid in the most of these existing works. In other words, the influence of the solid deformation on the meniscus shape and the liquid bridging force was not taken into account. It is noticed that the liquid bridging force and the solid deformation should be coupled for rigorous consideration of the problem. In some of the engineering applications, where the liquid bridging is not negligible, the coupling effect has been taken into account approximately by applying liquid bridging force as the total normal force in Hertz formula, then solve the implicit equation to get the contact radius and the liquid bridging force as functions of the filling angle and other geometrical and physical parameters (Section 3.3). In order to have a much more in-depth understanding of the contribution of the coupling effect, a FEM numerical simulations is carried out in present study.

In this research work, liquid bridging between a rigid sphere and an elastically deformable half space, as shown in Fig. 3.1a, is simulated numerically via FEA. An

arc of circle is used to model the meridian profile of a meniscus, which is called “circle approximation” by Orr et al [5]. They proved that when both filling angle and contact angle are small, solutions from “circle approximation” differ slightly from the exact ones. In our results, the dimensionless liquid bridging force and contact radius are plotted as functions of filling angle ψ , which measures the liquid volume, and a dimensionless parameter,

$$\chi = \frac{ER_0}{\gamma(1-\nu^2)}, \quad (3.1)$$

which measures the rigidity of the solid. In Eq. (3.1), E and ν are the elastic modulus and Poisson ratio of the half space, R_0 the radius of the rigid sphere, γ the surface tension of the liquid/air interface, respectively, as shown in Fig. 3.1b.

3.2 Liquid bridging induced contact

Let us consider an axial symmetric configuration where a rigid sphere is in contact with a linearly elastic half space without externally applied force. A droplet of liquid is presented at the interface and a liquid bridge with a concave meniscus is formed (as shown in Fig. 3.2a). A difference of hydrostatic pressure across the meniscus is produced, which is called Laplace pressure. The Laplace pressure and the surface tension in the meniscus are called liquid bridging forces, which pull the sphere and the half space together with a small contact area of radius c , as shown in Fig. 3.1b. The equilibrium is reached between the liquid bridging force and the contact pressure.

The Laplace pressure Δp across the liquid/vapor interface satisfies the following Laplace-Young equation (e.g. Orr et al, 1975):

$$\Delta p = 2H\gamma, \quad (3.2)$$

where $2H$ is the mean curvature of the meniscus, γ is the surface tension of the liquid in meniscus.

In Fig. 3.1b, θ_1 and θ_2 denote the contact angles at the two triple points with the sphere and the half space, respectively. R_0 is the radius of the sphere. ψ is filling angle that characterizes the volume of the small liquid drop. l is the outer radius of the half space/liquid interface.

Since the liquid bridging induced contact is only observed in micro-scale, the influence of gravity can be ignored and the mean curvature of meniscus is uniform and the pressure is same in the liquid. “Circle approximation” is used to approximate the meridian meniscus profile. Provided that the equation describing the half space surface after deformation under the liquid bridging force is $z = g(r)$, given θ_1 , θ_2 and ψ , by using “circle approximation”, the coordinate $(l, z_o = g(l))$ of the triple point on the half space and the meridian radius r_1 can be obtained from the following equations,

$$\begin{cases} \tan\{\arctan[g'(l)] + \pi/6\}(r-l) + g(l) = z \\ -\tan[\pi/2 - (\theta_1 + \psi)](r - R_0 \sin \psi) + R_0(1 - \cos \psi) = z \\ (r-l)^2 + (z - g(l))^2 = (r - R_0 \sin \psi)^2 + [z - R_0(1 - \cos \psi)]^2 \\ r_1 = \sqrt{(r - R_0 \sin \psi)^2 + [z - R_0(1 - \cos \psi)]^2} \end{cases} \quad (3.3)$$

where the point (r, z) is the curvature center of the meridian meniscus profile.

The second principal curvature must also be taken as constant because of constant mean curvature. A convenient and representative value of the second principal radius is the slant distance from the triple point on the sphere to the axis of symmetry as given by

$$r_2 = \frac{R_0 \sin \psi}{\sin(\theta_1 + \psi)}, \quad (3.4)$$

which is shown in Fig. 3.1b.

Eqs. (3.3) and (3.4) provide a good approximation for the mean curvature of $2H$ at small filling angles:

$$2H = 1/r_1 - 1/r_2, \quad (3.5)$$

where the mean curvature is positive when the meniscus is concave.

The total force caused by the liquid bridging between the sphere and half space consists two parts, namely, the surface tension force that resides in the meniscus and the capillary force due to Laplace pressure. The attractive force between the sphere and the half space is taken as positive.

The axial and radial components of surface tension force applied on the half space is

$$\begin{aligned} F_{sz} &= -F_s \sin(\theta_2 + \arctan[g'(l)]) = -2\pi l \gamma \sin(\theta_2 + \arctan[g'(l)]) \\ F_{sr} &= -F_s \cos(\theta_2 + \arctan[g'(l)]) = -2\pi l \gamma \cos(\theta_2 + \arctan[g'(l)]) \end{aligned} \quad (3.6)$$

The normal component of the capillary force applied on the half space is given by

$$F_{\Delta p} = \pi(l^2 - c^2) \times \Delta p \quad (3.7)$$

The total normal force in the axial direction is

$$F_{total} = |F_{sz}| + F_{\Delta p} = F_{\Delta p} + 2\pi l \gamma \sin(\theta_2 + \arctan[g'(l)]) \quad (3.8)$$

It is noticed that the contact radius c , the position of the half space surface after deformation $z = g(r)$, and the liquid bridging force are coupled together.

The Laplace pressure and surface tension are applied on the solids at the liquid/solid interface and solid/liquid/vapor triple point, respectively, as shown in Fig. 3.2a for the half space. These forces and pressure are applied on the surface of the elastic half space as the boundary conditions in our FEA calculation in Section 3.5.

3.3 The coupled and decoupled Hertz contact solutions

In many engineering applications, the above-mentioned liquid bridging contact configuration as shown in Fig. 3.1 has been simplified as a Hertz contact as shown in Fig. 3.3. In the simplified configuration, Fig. 3.3, the liquid bridging forces are summed up as F_{total} and considered as external force acting in the far field. The half

space surface outside the contact area is taken as a horizontal plane, and the contact radius is determined by Hertz contact solution as [34]

$$c = \left(\frac{3R_0 F_{total}}{4E^*} \right)^{\frac{1}{3}}, \quad (3.9)$$

where $E^* = E/(1-\nu^2)$ is the composite modulus (E and ν are the elastic modulus and Poisson's ratio of the half space, respectively). The liquid bridging force is, therefore, given by

$$F_{total} = \left(\frac{\cos(\theta_1 + \psi) + \cos\theta_2}{\sqrt{R_0^2 - c^2} - R_0 \cos\psi} - \frac{\sin(\theta_1 + \psi)}{R_0 \sin\psi} \right) \gamma \pi (R_0^2 \sin^2 \psi - c^2) + 2\pi R_0 \gamma \sin\psi \sin(\theta_1 + \psi) \quad (3.10)$$

It is noted that the contact radius (or contact area) and liquid bridging force are still coupled. Results of the contact radius and liquid bridging force by solving Eq. (3.9) and (3.10) simultaneously are called coupled Hertz approximate solutions in the present work.

If we ignore the coupling of c and F_{total} , the above-mentioned contact problem can be simplified. Setting $c=0$ in Eq. (3.10) to obtain F_{total} , then substituting F_{total} back into Eq. (3.9) to obtain c , F_{total} and c are called decoupled Hertz approximate solutions (Bhushan, 1999).

3.4 Dimensional analysis

The liquid-bridging force and contact radius of the liquid bridging-induced single asperity contact depend on a few governing parameters: the composite modulus E^* , the radius of the rigid sphere R_0 , the surface tension γ , the filling angle ψ , and the contact angles θ_1 and θ_2 . By applying dimensional analysis, we have

$$\begin{cases} \frac{F_{total}}{R_0\gamma} = f_m(\chi, \psi, \theta_1, \theta_2) \\ \frac{c}{R_0} = f_c(\chi, \psi, \theta_1, \theta_2) \end{cases} \quad (3.11)$$

where

$$\chi = \frac{E^* R_0}{\gamma} \quad (3.12)$$

is a dimensionless parameter.

For the coupled and decoupled hertz approximation solutions, by using the dimensionless quantities in Eq. (3.11), Eqs. (3.9) and (3.10) can be rewritten as

$$\frac{c}{R_0} = \left(\frac{3}{4} \times \frac{\gamma}{E^* R_0} \times \frac{F_{total}}{R_0 \gamma} \right)^{\frac{1}{3}} = \left(\frac{3}{4} \times \frac{1}{\chi} \times \frac{F_{total}}{R_0 \gamma} \right)^{\frac{1}{3}} \quad (3.13)$$

$$\frac{F_{total}}{R_0 \gamma} = \left(\frac{\cos(\theta_1 + \psi) + \cos \theta_2}{\sqrt{1 - \left(\frac{c}{R_0}\right)^2} - \cos \psi} - \frac{\sin(\theta_1 + \psi)}{R_0 \sin \psi} \right) \pi \left(\sin^2 \psi - \left(\frac{c}{R_0}\right)^2 \right) + 2\pi \sin \psi \sin(\theta_1 + \psi) \quad (3.14)$$

In the following numerical simulation, the half space is treated as linearly elastic and liquid contact angles θ are as fixed parameters. Our focus is placed on the parametric study of liquid bridging force and contact area as functions of dimensionless variables χ and ψ .

3.5 Finite Element Analysis

Due to the coupling effect between the liquid bridging and elastic deformation of the contacted half space, the contact radius, Laplace pressure and surface tension have to

be solved simultaneously. Firstly, we pick up an initial contact radius c_0 , the outer radius l_0 of liquid/half space interface, and the Laplace pressure and surface tension applied on the liquid/half space interface from the coupled Hertz solution (Section 3.3). Then, we set up a numerical model, on which the Laplace pressure and surface tension act. The numerical analysis provides us a new contact radius c_1 . c_1 is the desired contact radius if $\left| \frac{c_1 - c_0}{c_0} \right| < 10^{-3} \times (\text{the smallest size of the elements})$.

Corresponding l_0 and the applied forces are the desired. If not, an iteration procedure

is adopted as follows. If $\left| \frac{c_1 - c_0}{c_0} \right| \geq 10^{-3} \times (\text{the smallest size of the elements})$, Eqs. (3.2)

~ (3.6) are used to calculate the values of l_1 , the Laplace pressure and surface tension used in the second step of FEM simulations. The procedure is repeated until

$$\left| \frac{c_{i+1} - c_i}{c_i} \right| < 10^{-3} \times (\text{the smallest size of the elements})$$

The finite element code ANSYS 5.5 [45] is employed in the linear elastic analysis for the configuration shown in Fig. 3.1 and 3.2a. A sample of the FE mesh and boundary conditions are shown in Fig. 3.2b. The FE mesh consists of an elastic cylindrical solid with radius $5l$ and height $5l$ in normal contact with the surface of the rigid sphere on the bottom surface of this cylinder. The radius and height of $5l$ ensure that the influences of the two newly generated boundaries on the contact stresses and strains are small enough to be ignored. The surface of the rigid sphere is treated as a fixed contact boundary. The liquid bridging forces calculated from Eqs. (3.2), (3.5) and (3.6) act on the relative surface positions of the half space. The half space was constructed using two-dimensional 8 nodes axial-symmetric elements. Two-dimensional rigid-to-flexible surface-to-surface contact-pair elements are employed in the contact region between the rigid sphere and the half space. Knowing that the gradients of stresses and strains are high within the contact area, fine mesh is used for the possible contact region. A very fine mesh is also required in the region close to the edge of contact radius in order to obtain reliable values of the contact radius.

In our numerical computations, some material and physical properties are taken to be constant for the demonstration purpose. They are $\nu = 0.33$, $\gamma = 73 \times 10^{-6} \text{ N/mm}$ (water), and $\theta_1 = \theta_2 = 60^\circ$ (water with solids), respectively. The elastic modulus of the half space, the radius of the rigid sphere and the filling angle are given as follows for our parametric study.

Elastic modulus E : 1, 10 and 100 *Gpa*.

Radius of the rigid sphere R_0 : $10^{-4}, 10^{-3}, 3 \times 10^{-3}, 10^{-2}, 0.1$ and 1 *mm*.

Filling angle ψ : 1, 2, 4, 7, 10 and 15 *degree*.

The first two parameters are embedded in the dimensionless parameter

$$\chi = \frac{E^* R_0}{\gamma}.$$

Giving them separately here is meant to emphasis to the physical and geometric interpretation of the configuration and sense in real engineering applications.

Preliminary FEM analyses of two-dimensional and three-dimensional Hertz contact problems due to pressures applied far from the contact areas were done using similar model geometries and mesh densities, and same material properties as the above-mentioned ones. The results of the preliminary analyses were compared with the known analytical results from Hertz theory. The discrepancies of contact widths for 2-D Hertz contact problems were shown with a maximum of 0.03185%; for 3-D Hertz contact problems, the maximum of the discrepancies of contact radii was 0.02229%. It was shown that our numerical simulation produced results accurate enough to be used in the discussion in the next section.

The FEM numerical results show that the distribution of contact pressures is very close to a semi-ellipse, which is predicted by Hertz contact theory. Only a very small region close to the contact area has large stress and strain values. The region far from the contact area has low stress and strain values. The contact pressure can be very large, as high as 700 *Mpa*, due to the small contact area. This implies that the liquid bridging

induced contact is able to lead to non-negligible friction and wear between the surfaces in contact without any external load.

In the most of the cases, the contact region is continuous. However, in some rare cases, when the dimensionless parameter χ or the filling angle ψ is very small, the contact region becomes discontinuous. Those cases correspond to a very low elastic modulus with which the half space experiences large local deformation close to the regions the liquid bridging forces acting on. There is a ring of separation inside the contact region.

Our numerical results also show that the rotation angle at the triple point of the half space is smaller than 5 degrees. This indicates that the influence of the rotation angle on the Laplace pressure is small. However, when the triple point is near to the edge of contact area, the axial displacements of the half space surface increases greatly the values of Laplace pressure.

The dimensionless liquid bridging force and dimensionless contact radius are plotted as functions of the dimensionless parameter χ and filling angle ψ in Fig. 3.4 ~ 3.7. Analyses and graphing of the numerical data are performed by using the commercial software Origin 6.1 [46].

3.6 Discussion and summary

The finite element numerical simulation for the liquid bridging is carried out in the present study. It is noticed that the contact due to the liquid bridging force and deformation of the elastic half space is coupled in the FEA simulation. Based on our FEA numerical solution, the accuracies of the decoupled and coupled Hertz approximation solutions can be discussed.

The ratios of the decoupled and coupled Hertz solutions, force and contact area, to the FEA numerical solution are plotted in Fig. 3.8 and 3.9, where the values obtained via the approximated solutions are normalized by the FEA value. When the dimensionless

parameter χ and the filling angle ψ decrease, the errors of the Hertz approximate solutions (coupled and decoupled) increase dramatically. The errors of Hertz approximate solutions come from the fact that the liquid bridging force is assumed to act on the location far from the contact area, instead of acting on the ring area just outside the edge of the contact area. It is seen that Hertz contact solution substantially underestimates the contact area (Fig. 3.8) for low χ values. It is believed that this underestimation of contact area in turn gives underestimated friction force in the liquid bridging induced contact, which is commonly cited in literature of MEMS devices, such as hard-disk-drive.

Fig. 3.1a Liquid bridging between an asperity and a half-space

Fig. 3.1b The geometric parameters in liquid bridging contact

Fig. 3.2a The applied liquid bridging forces on the half space

Fig. 3.2b Axial symmetric FEM mesh for an elastic half space in contact with a rigid sphere

Fig. 3.4 Variation of dimensionless liquid bridging force with χ

Fig. 3.5 Variation of dimensionless liquid bridging force with ψ

Fig. 3.6 Variation of dimensionless contact radius with χ

Fig. 3.7 Variation of dimensionless contact radius with ψ

Fig. 3.8 The contact-area ratio of approximations to FEA solution

Fig. 3.9 The liquid bridging force ratio of approximations to FEA solution

Chapter 4

Energy Rate Consideration in Wafer Bonding

4.1 Introduction

Adhesive contact does not always just induce friction and wear, and therefore is not always considered as negative aspect in the contact. Wafer bonding technology based on the mechanism of adhesive contact has gained more and more attentions in industries of micro-systems, nonlinear optics, and light-emitting diodes, in which complex structures consisting of structured and plain silicon or other wafers can be simply joined to one mechanically stable configuration. With this technology, two free surfaces of mirror polished, flat and clean solids of almost any material, when brought into contact at room or elevated temperature, are locally bonded together to form a cohesive interface spontaneously as shown in Fig. 4.1, leading to electronic and opto-electronic devices of much superior performance [6, 17-22, 33]. Not like the conventional epitaxial growth technology, wafer-bonding technology can integrate dissimilar materials while generating few or no defects far from the bonded interface, except for a few atomic layers at the interface (e.g. the research work by Wada et al, 1993 [23]). Therefore, it is a promising and potential technology in semiconductor, electronics, and opto-electronics industries.

The micro-mechanism of wafer bonding comes from the short-range intermolecular and inter-atomic attraction forces, such as Van der Waals forces, which are one or two orders of magnitude lower than typical values for covalent bonding [6, 21]. In macroscopic viewpoint, this phenomenon can be explained by the reduction of surface energy after bonding [33]. Provided the sum of the two surface tensions exceeds the interface tension (This can always be satisfied after surface treatment [33]), when two surfaces join to form an interface, the net free energy reduces. The reduction of energy per unit area, known as the work of adhesion, is

$$\Gamma = \gamma_1 + \gamma_2 - \gamma_{12}, \quad (4.1)$$

where γ_1 and γ_2 are the surface tensions of two wafer materials respectively, and γ_{12} is the interface tension [7]. It is Γ to motivate bonding, therefore, it can be considered as driving force for the wafer bonding process in the absence of external pressure.

It is noticed that wafer surfaces always possess certain geometrical (e.g. rough surface considered by Yu and Suo, 1998 [33]) or physical mismatch (e.g. lattice mismatch considered by Liao, 1997 [19]). Two wafers are actually in micro-contact at asperities. When the wafer surfaces join to form an interface in the absence of large external pressure, the mismatch has to be accommodated by elastic distortion, plastic deformation, or mass transport. Resistance exists during the formation of an interface. Therefore, the bonding can only proceed when the driving force, in addition to external work, is high enough to overcome the resistance. Whether or not two wafers bond depends on the driving force and the roughness and waviness of the two wafers [6].

Based on the energy concept, many models have been proposed to explain the micro-scale mechanism of the wafer bonding. Stengl *et al* [30] proposed the gap-closing model. Tong and Gösele [21, 31, 32] then developed this gap-closing model further by considering the effect of elasticity on wafer bonding. They gave expressions of complete bonding conditions in the limits of very thin and very thick wafers, where the elasticity theory was used to study the energy balance between the released surface energy during bonding and the installed energy due to the elastic distortion of the wafer. Yu and Suo [33] analyzed the model of Tong and Gösele in full ranges of parameters (i.e., the thickness and the composite Young's modulus of the wafers, and the amplitude and wavelength of the misfit gap) and presented a wafer bonding criterion, which they demonstrated by an analytical solution for the three-dimensional elastic field induced by the adhesive complete contact between two wavy sinusoidal wafer surfaces. Based on the energy balance between the strain energy installed in the elastic bodies and the released surface energy, they gave the critical condition for complete wafer bonding. Yu and Suo's model results in the same results as the gap-closing model. Liao [19] described a model for calculating the elastic strain/stress field near wafer fusion interface due to lattice mismatch (i.e., misfit dislocations)

alone. It gave a reasonable stress field at a distance comparable to the dislocation spacing, but inapplicable near the dislocations. He did not consider the geometric misfit between the surfaces, and the critical condition for bonding. Gui et al [22] studied the effect of random surface roughness on direct wafer bonding based on DMT theory [47, 48]. Since DMT theory is only applicable to cases with small contact radius, which cannot take into account the interactions among asperities, Gui *et al*'s results for complete bonding and partial bonding with large contact area definitely led to large error.

It is noticed that the gap-closing model and Yu and Suo's model were concentrated only on the completely bonding (bonding over the entire area) and based on the criterion (hereafter we call it *total energy difference criterion*),

$$\Gamma A_{\text{total}} > U_{\text{total}}, \quad (4.2)$$

where A_{total} is the total interface area and U_{total} is the total strain energy installed in the bi-material after the external force is removed. Yu and Suo [33] used Eq. (4.2) for examining the bonding of two wavy surface wafers. However, this criterion based on the difference between the total energy before and after bonding may not be able to explain certain phenomena, such as partial bonding, external loading assisted bonding, and measurement of the bonding strength of the interface, observed in the experiments [6].

It has been agreed that the wafer bonding can be considered as an inverse process of crack propagation [7, 49]. Thus, the Griffith energy release rate criterion [50] for determining the propagation of crack can be extended to the wafer bonding process for explaining the above-mentioned experimental observations that have not been covered by the total energy difference criterion. Our understanding at present moment is that the energy rate concept determines whether the wafers can be bonded together and provides the detailed history of the bonding process, while the energy based criterion, for example Eq. (4.2), is only a necessary condition for the wafers to be bonded together. The statement is visualized in Section 4.3 to 4.6 via finite element numerical simulations.

In the following sections, we revisit the wafer-bonding criterion for rough surfaces. The resistance to wafer bonding, namely, the released surface energy required to form an unit area of interface (*energy absorption rate*), is defined. A new criterion based on energy absorption rate (*energy absorption rate criterion* called in this work) is proposed in Section 4.2. In Section 4.3, a FEM numerical model for the bonding between a surface with sinusoidal profile and a flat half-space was set up for the demonstration purpose of this new criterion. In Sections 4.4, the self-activated bonding is investigated by using the energy absorption rate criterion. The function of external loading to wafer-bonding process is investigated in Section 4.5. The parametric study for the effects of the surface shape and plasticity of the wafer materials are considered in Section 4.6. Some discussions on the application of the energy absorption rate criterion are given in Section 4.7. The research work is summarized in Section 4.8.

4.2 Energy absorption rate consideration in wafer bonding

In the well-known Griffith criterion [50] for predicting crack propagation, the energy release rate from an elastic-plastic body must be larger than that needed to form unit area of crack surface. As the wafer bonding process can be considered as an inverse process of crack propagation [7, 49], we hereby propose an energy rate based criterion for wafer bonding.

For wafers to bond at the room temperature, in the absence of large external pressure, the misfit gap between the wafers must be accommodated by elastic or elastic-plastic deformation. Fig. 4.1 illustrates this problem. Prior to contact, two wafers are not perfectly flat. Once brought together, they contact at a few points first (Fig. 4.1(a)). In this state, the total free surface area is large, and so is the free energy. To reduce the free energy, the wafers deform elastically or elastically-plastically and transform the free surfaces into an interface. If the misfit is large, the wafers absorb too much energy and the resistance to bonding is large, and the wafers will bond only over patches of the surface area, leaving the rest of the area unbonded (Fig. 4.1(b)). If the

misfit is small, the wafers absorb a small amount of energy and the resistance to bonding is also small, and the wafers will bond over the entire area (Fig. 4.1(c)) [33].

Let us consider the quasi-static process of wafer bonding with external loading. For an infinitesimal increase of the interface area, δA , the reduction of surfaces energies $(\gamma_1 + \gamma_2)\delta A$, the increase of interface energy $\gamma_{12}\delta A$, the work generated by external loading δW , and strain energy δU which is generated near interface owing to mismatch of geometric configuration and material properties between two wafers, satisfy the following equation,

$$(\gamma_1 + \gamma_2 - \gamma_{12})\delta A + \delta W - \delta U = \Gamma \delta A - \delta(U - W) \geq 0. \quad (4.3)$$

It is straightforward to identify that *driving force* to increase the interface area is Γ and the *resistance* to form the interface is,

$$R = \frac{\partial(U - W)}{\partial A}, \quad (4.4)$$

which is the absorbed surface energy in forming unit area of the interface, or *energy absorption rate*. This concept can be applied for either elastic or elastic-plastic deformations.

With the definition of resistance, Eq. (4.4), Eq. (4.3) is read as:

$$\Gamma \geq R, \quad (4.5)$$

which is called *energy absorption rate criterion*. If the present contact area is A , and a far field pressure acts on the wafer, the energy absorption rate criterion indicates the following seven possibilities in a wafer bonding process.

(1) *In a bonding process*

In this case, the resistance R is smaller than the driving force Γ . Therefore, to reduce the energy of the whole system, the bonding process proceeds and the interface area increases until complete bonding.

(2) *In a de-bonding process*

In this case, the resistance R is larger than the driving force Γ . Therefore, to reduce the energy of the whole system, the two wafers are in a de-bonding process and the interface area reduces until to 0.

(3) In a steady equilibrium state

In this case, the resistance R is equal to the driving force Γ . However, R is smaller than Γ for an infinitesimal decrease of the interface area and is larger than Γ for an infinitesimal increase of the interface area. The bonded wafers are in a steady equilibrium state. An infinitesimal departure from the present state makes the system go back to the present state. A larger far field pressure is needed for the interface area to increase.

(4) In an unsteady equilibrium state: A1

In this case, the resistance R is also equal to the driving force Γ . However, R is larger than Γ for an infinitesimal decrease of the interface area and is smaller than Γ for an infinitesimal increase of the interface area. The bonded wafers are in a unsteady equilibrium state. An infinitesimal departure from the present state makes the system go far from the present state. That is, an infinitesimal increase of the interface area makes the system go into a bonding process, vice versa.

(5) In an unsteady equilibrium state: A2

In this case, the resistance R is equal to the driving force Γ in a infinitesimal domain near the present state. The bonded wafers are in a unsteady equilibrium state. An infinitesimal departure from the present state makes the system go to the new unsteady equilibrium state similar as the present state.

(6) In an unsteady equilibrium state: A3

In this case, the resistance R is equal to the driving force Γ . However, R is larger than Γ in a infinitesimal domain near the present state. The bonded wafers are in a unsteady equilibrium state. A negative infinitesimal change of the interface area makes the

system go into a de-bonding process. But a positive infinitesimal change of the interface area makes the system go back to the present state.

(7) In an unsteady equilibrium state: A4

In this case, the resistance R is equal to the driving force Γ . However, R is smaller than Γ in a infinitesimal domain near the present state. The bonded wafers are in a unsteady equilibrium state. A negative infinitesimal change of the interface area makes the system go back to the present state. But an positive infinitesimal change of the interface area makes the system go into a bonding process.

The energy absorption rate criterion and the total energy difference criterion give different predictions to the final state of two bonded wafers. For example, Fig. 4.2 describes four possible relations between the resistance and the work of adhesion. Although the total energy difference criterion might explain Case 1 and 4, it cannot be used to investigate Case 2 and 3. For example, for Case 2 in Fig. 4.2(a), the shadow areas below and above the curve Γ are equal to each other, according the total energy difference criterion, the interface area should be A_2 , but it is A_1 according to the energy absorption rate criterion. For Case 3 in Fig. 4.2(b), things are similar.

It is noticed that an integration of Eq. (4.3) with respect to the area A leads to Eq. (4.2), which has a same form as the total energy difference criterion.

$$\Gamma A > U. \quad (4.6)$$

For wafer bonding, Eq. (4.6) is only a necessary condition, while Eq. (4.3) (or Eq. (4.5)) is a sufficient and necessary condition.

In order to give a quantitative description on this problem, we need to have numerical results from a finite element simulation for a wafer bonding process as follows.

4.3 A finite element method simulation for bonding between elastic wavy surfaces

As mentioned in previous section, the driving force of wafer bonding, Γ , between a given material pair is completely determined by the material properties, while the resistance is also associated with the shape of the two surfaces and the external loading. The relationship between them is the key to analyze the process of wafer bonding. Yu and Suo [33] obtained the analytical results in the case of two-dimension periodic problem, in which only the final stage was considered without the whole process of the bonding. When only the elastic bonding process is considered without external loading, by introduction of periodic distributions of infinitesimal climb edge dislocations to treat the closing of the initial gaps, the elastic energy in the wafers can be calculated by a straightforward integration without need of numerical calculations. However, it would not work and this boundary value problem has no analytical solution for the bonding process with plastic deformation and external loading. Thus, in the present study, a finite element analysis for the whole bonding process, from zero contact to full contact, is carried out. The strain energy including plastic work and resistance R are calculated from the numerical stress field for analyzing the whole bonding process in the following sections.

In the frictionless contact of elastic solids, the contact stresses depend only upon the relative profile of their surface. Therefore, the system can be replaced by a flat rigid surface in contact with a body (having an effective modulus E^* with $\frac{1}{E^*} = \frac{1-\nu_1^2}{E_1} + \frac{1-\nu_2^2}{E_2}$ and a yielding stress σ_s). The surface profile results in the same un-deformed gap between the surfaces. With this in our mind, we consider the bonding between a wavy surface with periodic profile and a rigid flat surface as shown in Fig. 4.3. The profile of the wavy surface is described by

$$H(x) = 2\Delta[\sin^2(\frac{\pi x}{\lambda}) + \alpha \sin^2(\frac{2\pi x}{\lambda})], \quad (4.7)$$

where λ is the wavelength, and 2Δ the amplitude. In the figure, for clarity, the ratio $2\Delta/\lambda$ has been greatly exaggerated. The parameter α is introduced to investigate the effects of the profile shape on wafer bonding in Section 4.6.1.

Owing to the periodicity, we only considered one wavelength as shown in Fig. 4.3. Along the left and right boundaries, the periodic constraints are applied, that is u_x are same at $x = \pm \lambda/2$, respectively. To calculate the resistance of wafer bonding, the displacement boundary condition is applied on the wavy surface to have a contact width $2a$. The pressure, p , acts in far field of the wafer body.

The commercial finite element package ANSYS [45] was employed to carry out the elastic-plastic numerical calculation. In the FE model, the value of the wafer thickness, t , is taken to be 10 times of λ . It is large enough to ensure that effect of the contact between the wafer and the rigid plane on the stress field far from the contact interface can be ignored. The wafer was constructed using PLANE42 (two-dimensional structural solid) elements. The field of displacements, strains and stresses are calculated for the given contact width, $2a$, increasing from 0 to λ . The strain energy including plastic work installed in the solids, U , and the work applied by the far field pressure, W , can be obtained. In turn, the resistance R , defined by Eq. (4.4), is obtained.

$$R(2a) = \frac{\partial(U - W)}{\partial(2a)}. \quad (4.8)$$

For this elastic-plastic boundary value problem, all the independent geometrical and physical quantities are taken as follows:

- (i) The geometrical dimensions: λ (asperity wavelength), Δ (asperity amplitude), α (indicating the asperity shape), a (half contact width);
- (ii) The material properties: E (Young's modulus), ν (Poisson's ratio), σ_s (yielding stress) of the wafer.
- (iii) The far field pressure: p .

Dimensional analysis showed that there are five independent dimensionless quantities for this problem, i.e., $\frac{\Delta}{\lambda}, \frac{\sigma_s \lambda}{E^* \Delta}, \alpha, \frac{p \lambda}{E^* \Delta}, \frac{2a}{\lambda}$. We choose the geometrical and physical quantities for the numerical simulation as follows:

$$\begin{aligned} \lambda &= 2\text{mm}; t = 20\text{mm}; E = 50\text{GPa}; \nu = 0.3; \\ \Delta &= 0.005, 0.0025, 0.0005\text{mm}; \\ \sigma_s : \frac{(1-\nu^2)\lambda}{E\Delta} \sigma_s &= \frac{(1-0.3^2) \times 2\text{mm}}{50\text{GPa} \times 0.005\text{mm}} \times \{20\text{MPa}, 200\text{MPa}, 500\text{MPa}, 1\text{GPa}, \infty\} \end{aligned}$$

The material is elastic-perfectly-plastic. It is elastic when σ_s is taken to be ∞ .

Accordingly, the dependent quantities to be presented as results are also dimensionless. Here we introduce a quantity that has same dimension as the resistance:

$$R^* = \frac{\pi E^* \Delta^2}{2 \lambda}. \quad (4.9)$$

The stress and strain in the wafer scale as $E^* \Delta / \lambda$ and Δ / λ respectively. Therefore, the density of strain energy scales as $E^* (\Delta / \lambda)^2$. The average strain energy made over the wavelength is defined by $\bar{U} = U / \lambda$ in the wafer of unit thickness, which has the same dimension as R^* .

The finite element numerical results shown in Fig. 4.4 quantitatively indicate the variation of $\bar{U}$ and R with respect to $2a/\lambda$. The important and detailed features in Fig. 4.4 are as follows. (1) The resistance $R = 0$ when the contact area equals to zero ($a = 0$). (2) The resistance R increases as the contact area increases and reaches its maximum value $R_m = 1.02R^*$ at $2a_m / \lambda = 0.68$. (3) The resistance decreases to zero when the interface is completely bonded.

The numerical simulations showed that the effect of Δ/λ on the dimensionless strain energy and resistance is ignorable.

Our FEM numerical results are verified for the complete bonding of the elastic wafer in Appendix. The strain energy and the maximum normal stress on the interface are compared with the corresponding analytical ones. It is shown that our numerical simulation produced results accurate enough to be used in the discussion of the wafer bonding process.

Since the resistance varies with the contact area, it may not be proper to analyze the wafer bonding process by the total energy difference criterion, especially when the driving force of wafer bonding is less than the maximum resistance. In the following sections, by using the finite element numerical results, we analyze the process of the self-activated bonding and the external loading assisted bonding, and discuss the effects of the asperity shape and plasticity on wafer bonding.

4.4 Self-activated wafer bonding

When the wafers are free of external loading, the wafers are only bonded due to the work of adhesion. This is called self-activated wafer bonding in our work. The dimensionless strain energy and resistance are presented as functions of the contact area $2a/\lambda$ in Fig. 4.4. Here, only elastic wafers with a surface profile of $\alpha = 0$ are considered. In this section, with the finite element results in Fig. 4.4, we have quantitative description on various bonding phenomena by using the energy absorption rate criterion, which is different from the prediction by using the total energy difference criterion, as follows.

4.4.1 Self-activated complete bonding

According to the energy rate consideration, when the driving force Γ is greater than the maximum resistance $1.02R^*$, the wafer bonding is self-activated without any external loading. For this case,

$$\frac{\Gamma\lambda}{E^*\Delta^2} > 1.60 \quad \text{or} \quad \Gamma/R^* > 1.02. \quad (4.10)$$

The strain energy in the wafer is $\bar{U}(1)/R^* = 0.50$.

In order to have a qualitative measurement for the bonding strength of the bonded interface, the *specific effective bonding energy*, E_b , which is the external mechanical work needed for separating per unit nominal area of contact interface, is defined.

$$E_b = \Gamma - \bar{U}(1). \quad (4.11)$$

Therefore, for the self-activated complete bonding, we have

$$E_b = \Gamma - \bar{U}(1) > (1.02 - 0.50)R^* = 0.52R^*. \quad (4.12)$$

4.4.2 Self-activated partial bonding

When Γ is less than the maximum resistance $1.02R^*$, only partial bonding can be reached via the self-activated mechanism. The contact area is determined by

$$R(2a') = \Gamma, \quad (4.13)$$

where $a' = a / \lambda$. There are two solutions to this condition. They are denoted by $2a_1'$ and $2a_2'$ ($a_1' < a_2'$) respectively. Only the smaller, $2a_1'$, can be reached spontaneously. Although further bonding is allowed by the energy-based consideration, the resistance R is larger than the driving force Γ after this point.

In this case, the specific effective bonding energy is

$$E_b = 2a_1' \Gamma - \bar{U}(2a_1'), \quad (4.14)$$

which is always positive.

4.4.3 Prediction based on the total energy difference criterion

As mentioned in Section 4.1, the total energy difference criterion has been used to predict the complete bonding by former researchers. According to this criterion, the necessary and sufficient condition for complete wafer bonding is

$$\Gamma\lambda > U(\lambda). \quad (4.15)$$

In a dimensionless format, as shown in Fig. 4.4, the above equation can be rewritten as

$$\frac{\Gamma\lambda}{E^*\Delta^2} > 0.783, \text{ or } \Gamma/R^* > 0.5. \quad (4.16)$$

This dimensionless parameter was used by Yu and Suo [33] and Gui *et al* [22]. It is listed here for easy comparison with our quantitative description of wafer-bonding process by using the energy absorption rate criterion. According to the total energy difference criterion, if and only if Eq. (4.16) is satisfied, it is claimed that the wafers are bonded completely.

In this case, the specific effective bonding energy is

$$E_b = \Gamma - \bar{U}(1) > 0. \quad (4.17)$$

4.4.4 Comparison of the two criteria

Comparing Eq. (4.16) with Eq. (4.10), we conclude that the energy absorption rate criterion describes a more rigorous condition for the self-activated wafer bonding. In other words, some of the bonding claimed by Eq. (4.16) cannot be reached. Flatter surface, softer materials and higher interface driving force are needed. It is also noticed that the specific effective bonding energy predicted by the energy absorption rate criterion has a minimum $0.52R^*$, but it is 0 predicted by the total energy difference criterion.

From the numerical simulation, we realized that the total energy difference criterion does not provide the information during the bonding process, and it predicts a value of the work of adhesion smaller than the real value. It assumes that the deformed wafers absorb all the released surface energy when two surfaces are bonded to an interface. It overlooks the variation of the resistance with the contact area. The description of the bonding details during the process relies on the energy rate consideration, where competition between driving force, Γ , and resistance, R , introduces the external loading into the picture (as discussed in the next section).

4.5 External loading assisted wafer bonding

When Γ is less than $1.02R^*$, only partial bonding can be reached via the self-activated mechanism, which only drives the bonding interface to $2a_1'$, as stated in Section 4.4.2. Therefore, an external loading is needed to reduce the resistance to the level smaller than the driving force Γ , and then the bonding has the potential to be strengthened.

With the increase of external loading, the width of contact increases. Even when the contact area is larger than $2a_u'$ (at which the resistance is the maximum for the self-activated bonding), external loading is still needed since the driving force is less than the resistance. Before the contact area reaches $2a_2'$, the contact area above $2a_1'$ will be released if the external force is removed from the wafers. The external load is needed until the contact area (bonded area) reached the value of $2a_2'$, when the self-activated mechanism prevail as the driving force is larger than the resistance again (as shown in Fig. 4.5(d)). The wafer surfaces are bonded together spontaneously until the whole area is in contact, and the complete bonding is reached.

When the contact area increases from $2a_1'$ to $2a_2'$, the external loading reduces the resistance to a level not larger than the driving force Γ . If a pressure acts on the far field of the wafer to press the wafers into a bonding process, the minimum of this pressure, p , satisfies

$$R(2a) = \frac{\partial(U(2a) - W(2a))}{\partial(2a)} = \Gamma, \quad (4.18)$$

where $R(2a)$, $U(2a)$ and $W(2a)$ are the relative values for a contact width $2a$ under the far field pressure. Under p , the wafers enter a quasi-static bonding process. Fig. 4.5 and Fig. 4.6 present the pressures, the relative far field displacements normal to the interface, the strain energies, and resistances for $\Gamma = 0.42$ and $\Gamma = 0.74$ respectively. With the contact area increasing from $2a_1'$ to $2a_2'$, the far field pressure increases from 0, then attains its maximum, and finally come back to 0 again. As the far field pressure applies to the wafer, in domain $(2a_1', 2a_2')$, the strain energy is larger than

that corresponding to the bonding process without external loading, and the resistance is maintained at Γ .

When the wafers are completely bonded due to both the self-activated bonding and the external loading assisted bonding, the complete bonding state is maintained, except that the interface is separated into two surfaces by an external pull force. Fig. 4.7 shows the energy release rates G vs. free area A_{free} when the interface is separated from the completely bonded interface at four different positions. The energy release rate tends to 0 when the free area tends to 0 for all the four positions. The position with largest slope in Fig. 4.7 can be separated most easily. For a non-zero value of the work of adhesion, de-bonding phenomenon starting from the completely bonded wafers does not happen without the assist of external pull force. Thus, the external work applied by p is locked near the interface. That is one of the reasons that larger external force is helpful in forming the bonding interface.

In this case, the specific effective bonding energy is also given by Eq. (4.11). However, it can be both negative and positive, and this depends on the value of the work of adhesion of the interface.

The total energy difference criterion cannot predict the external loading assisted bonding phenomenon, as it does not taken into account the function of external loading.

4.6 Effects of asperity shape and plasticity on wafer bonding

There are some other parameters affecting the process of the wafer bonding. Among the many parameters, we would like to discuss the following two geometric and material factors, namely, the asperity shape on the surface and the plastic deformation in the wafers.

4.6.1 Effects of the asperity shape

To change the asperity shape on the contact surface, we can use various α values in Eq. (4.16). Fig. 4.8 shows that for “fatter” (smaller α) and “slimmer” (larger α) asperities, which represent different wafer surfaces. Figs. 4.9 and 4.10 show the effects of α on the strain energy and resistance, respectively. It shows that even with the same wavelength and the same height, the asperity shape on the surface plays a role in the wafer bonding process.

In Fig. 4.9, the strain energy for a surface with slimmer asperities is larger than that for fatter ones in the partial bonding process. Nevertheless, when complete bonding, the difference reduces and the values of the strain energies are very close to each other for different asperity shapes.

On the other hand, the asperity shape does play a role in the maximum resistance of wafer bonding as shown in Fig. 4.10. The maximum resistance is larger for a surface with “fatter” asperities. Therefore, it is harder to bond wafers with “fatter” asperities. This indication should be meaningful even for randomly distributed rough surface, although the current conclusion is obtained from a periodical distributed rough surface.

4.6.2 Effects of plastic deformation

Plastic deformation occurs when the loading is high especially for the materials with low yielding stress and full range of contact, such as wafer bonding. It is intuitive concept that the soft materials (low yielding stress) provide low resistance to the bonding. The quantitative consideration is given in Fig. 4.11 and Fig. 4.12.

For the complete bonding, the maximum normal stress on the interface within elastic range is estimated by (see Appendix)

$$\sigma_z = \frac{\pi E^* \Delta}{4\lambda}. \quad (4.19)$$

Therefore, $\sigma_y \lambda / (E^* \Delta)$ is taken as a dimensionless parameter that describes the level of the yielding in our data presentation. In Fig. 4.11 and Fig. 4.12, three curves are

shown for various deformation situations. For elastic deformation, $\sigma_s \lambda / (E \Delta)$ value is infinitely large, while for mild plastic deformation the value is taken 1.456, and for deep plastic deformation the value is taken as 0.1456. The quantitative study shown in Fig. 4.11 and Fig. 4.12 confirmed our intuitive concept that the low yielding stress materials are much easier to be bonded as the resistance is low.

4.7 Discussion on the application of the energy absorption rate criterion

The energy absorption rate criterion discussed in the present work does not specify the type of bonding. Heat treatment and chemical treatment can be used to improve the surface property and increase the work of adhesion of the contact interface [33]. The effects of physical and chemical changes to the wafer surfaces enter the criterion through the work of adhesion.

We can use a softer wafer material or a flatter wafer surface to get a better bonding property. The wafer surfaces can be modified by means of chemical mechanical polishing (CMP) and chemical etching [22].

We demonstrate the energy absorption rate criterion in the present work by using a wafer with a periodical wavy surface, which has only one wavelength and amplitude of the asperities. In practice, the wafer surface contains many wavelengths, each having its own amplitude. If one wavelength and amplitude are dominating, the criterion in the present work can be used to the wafer-bonding problem directly; If not, a Fourier transform can be performed to obtain the asperity amplitude as a function of the asperity wavelength [33]. Then the absorbed strain energy in wafer bonding process for each wavelength can be obtained by considering the wafer surface as a periodical wavy surface, and the total absorbed strain energy in the bonding process can be obtained by linear superposition as a function of the real contact area. The total absorbed strain energy can then be used to obtain the relation of the resistance as a function of the real contact area. However, this procedure is much harder and

unrealistic in practice. Therefore, we can treat the wafer-bonding problem approximately as follows. With the asperity amplitude as a function of the asperity wavelength from the Fourier transform, we can obtain the wavelength and amplitude of the wafer that are most difficult to bond. The present criterion can be used with these amplitude and wavelength as the geometrical parameters of the wafer.

4.8 Concluding remarks

In the present work, we considered the wafer bonding process via an energy absorption rate concept. Whether or not two wafers bond depends on the competition between the driving force (the work of adhesion) and the resistance to bonding. When the driving force is larger than the resistance, the bonding process proceeds; otherwise, the system enters a de-bonding process. The wafer bonding can be divided into three categories, i.e., the self-activated complete bonding, the self-activated partial bonding, and the external loading assisted bonding.

For two elastic wafers to be bonded, the energy absorption rate criterion gives the work of adhesion of the interface in terms of the wafer material properties, the surface flatness, and shape of the surface asperity. It is appropriate to have a large work of adhesion of the wafer interface, small Young's modulus of the wafer materials, and flat surface.

With the interface area increasing from zero to the entire wafer surface, the strain energy monotonically increases, but the resistance increases from zero firstly, then reaches the maximum, and finally decreases to zero.

The self-activated complete bonding can be achieved only when the driving force is larger than the maximum resistance. The energy absorption rate consideration gives a more rigorous condition than the total energy difference criterion for this bonding type.

If the driving force is smaller than the maximum resistance, the self-activated partial bonding is achieved with the resistance equal to the driving force.

When the maximum resistance is large and the self-activated complete bonding cannot be reached, an external loading can reduce the resistance to a level smaller than the driving force. Therefore, two wafers can still be bonded completely, and the complete bonding state is maintained. The external work applied in the bonding process is stored near the interface.

Plasticity can produce a large reduction to the resistance. The low yielding stress materials are much easier to be bonded.

There is a new parameter defined in Section 4.4.1, namely, the specific effective bonding energy. It is an important parameter for characterizing the bonded interface. With this parameter, we not only understand the wafer bonding process, but also know the bonding strength of the interface.

There are many interesting topics related to the rough surface bonding for further research. The random rough surface may show some new phenomena that have not been observed in our periodical wavy surface configuration. The existing contact mechanics treats the contact among the asperities as independent since the total contact area is far from the full contact. The wafer bonding, as a real engineering application, initiates a new research direction for the contact mechanics where asperity contacts are coupled, as the contact area is close or equal to the full contact. We have proposed a self-consistent scheme for the random rough surface in full contact [51], which can be incorporated into the further research.

(a)

(b)

(c)

Fig. 4.1 Schematics of wafer bonding

(a) Initial contact (b) partial bonding (c) complete bonding

Fig. 4.2 Schematic show of formation of interface

Fig. 4.3 The configuration used in FEA calculation

Fig. 4.4 Normalized strain energy and energy absorption rate versus normalized contact area

Fig. 4.5 External loading reduces the resistance for $\Gamma/R^*=0.42$

(a) far field pressure vs. contact area; (b) far field displacement vs. contact area; (c) far field pressure vs. far field displacement; (d) strain energy and resistance vs. contact area

Fig. 4.6 External loading reduces the resistance for $\Gamma/R^*=0.74$

(a) far field pressure vs. contact area; (b) far field displacement vs. contact area; (c) far field pressure vs. far field displacement; (d) strain energy and resistance vs. contact area

Fig. 4.7 Energy release rates G vs. free area A_{free} when the interface is separated from the completely bonded interface at four positions

Fig. 4.8 Shapes of wavy surfaces with different α

Fig. 4.9 Variation of normalized average strain energy vs. contact area with different α

Fig. 4.10 Variation of resistance vs. contact area with different α

Fig. 4.11 Strain energy vs. contact area with different yielding stresses

Fig. 4.12 Resistance vs. contact area with different yielding stresses

Chapter 5

Summary and Conclusions

The adhesive micro-contact is induced by the attraction forces between contact surfaces, i.e., the liquid bridging force and other adhesive forces between solids. In the present thesis, we focus on two aspects: the research on the liquid bridging induced contact and the research on the energy absorption rate criterion for wafer bonding from the point of view of micro-mechanics. Three problems related to adhesive micro-contact were analyzed. Firstly, the evolution process of a liquid bridge between a rigid cylinder and a rigid plane was investigated by setting up a theoretical model based on the frame of irreversible non-equilibrium thermodynamics. Secondly, the coupling effect between the solid deformation and the liquid bridge in the liquid bridging induced contact was analyzed numerically by finite element method (FEM). Finally, motivated by the Griffith energy release rate criterion for predicting crack propagation, the energy absorption rate criterion was proposed to predict the wafer bonding process. In this chapter, the main contributions are summarized.

For the evolution process of a liquid bridge between a rigid cylinder and a rigid half space, a theoretical model was set up based on the frame of irreversible non-equilibrium thermodynamics to investigate the kinetic evolution process of the liquid bridge. The ingredients of the non-equilibrium thermodynamics for this evolution process, i.e., the free energy of the system, the motions of the meniscus and the two triple point lines (TPLs), the driving force of the evolution, and the kinetic laws between the actual velocities and the driving forces, were formulated. By applying the weak statement to these ingredients, a set of nonlinear ordinary differentiation equations (ODEs) with generalized coordinates was obtained, which govern the dynamical evolution process. Numerical simulations for the set of ODEs were carried out and the effects of the control parameters on the evolution process were discussed.

From our theoretical model, the evolution process of a liquid bridge between a rigid cylinder and a rigid plane is controlled by 4 dimensionless control parameters, i.e., the initial average filling angle ψ_0 , the equilibrium contact angle θ , the dimensionless mobility, and the dimensionless wavelength of the liquid bridge, $\lambda/(R\psi_0)$. Our numerical results show that ψ_0 and $\lambda/(R\psi_0)$ have very small effects on the normalized sizes of the liquid bridge and the normalized free energy of the system. The evolution process is more rapid for a liquid bridge with a smaller contact angle. A large time is needed for a liquid bridge with small mobility $\tilde{M}$ to evolve to the final equilibrium configuration. Our numerical results show that the liquid droplets between contact surfaces will evolve to an equilibrium liquid bridge

For the coupling effect between the solid deformation and the liquid bridge in the liquid bridging induced contact, a FEM numerical simulation for the adhesive contact between an elastically deformable half space and a rigid sphere was carried out. The rigorous description of the half space surface under deformation was achieved. It is noticed that the contact due the liquid bridging force and deformation of the elastic half space is coupled in the finite element analysis (FEA) simulation. A dimensional analysis reveals that this problem is determined by two governing parameters, i.e., χ (measurement of relative stiffness of the contact bodies) and the filling angle ψ (measurement of liquid volume). Parametric studies reveal the dependence of the contact area and the liquid bridging force on the filling angle and the dimensionless group χ .

Based on our FEA numerical solution, the accuracies of the decoupled and coupled Hertz approximation solutions were discussed. When the dimensionless parameter χ and the filling angle ψ decrease, the errors of the Hertz approximate solutions (coupled and decoupled) increase dramatically. The errors of Hertz approximate solutions come from the fact that the liquid bridging force is assumed to act on the location far from the contact area, instead of acting on the ring area just outside the edge of the contact area. It is seen that Hertz contact solution substantially underestimates the contact area for low χ values. It is believed that this underestimation of contact area in turn gives underestimated friction force in the liquid bridging induced contact, which is

commonly cited in literature of micro-electro-mechanical systems (MEMS) devices, such as hard-disk-drive.

Wafer bonding used in MEMS is a technology based on the mechanism of adhesive contact. Motivated by the energy release rate criterion for predicting crack propagation, we proposed an energy absorption rate criterion for analyzing the process of wafer bonding. We demonstrated it by discussing the bonding process between a rigid plane and an elastic-plastic wafer with a periodical wavy surface.

Our research showed that whether or not two wafers bond depends on the competition between the driving force (the work of adhesion of the contact interface) and the resistance to bonding. Only when the driving force is larger than the resistance, the bonding process proceeds. In the bonding process, the resistance increases from zero firstly, then reaches the maximum, and finally decreases to zero. Therefore, wafer bonding can be divided into three categories, i.e., self-activated complete bonding, self-activated partial bonding, and external loading assisted bonding. In an external loading assisted bonding process, the external loading reduces the resistance to a level smaller than the driving force. The external work applied in the bonding process is locked near the interface. Plasticity can produce a large reduction to the resistance. The low yielding stress materials are much easier to be bonded.

List of Publications

- [1] “Elastic analysis for liquid-bridging induced contact”, Accepted for publication in *Finite Elements in Analysis and Design*.
- [2] “Energy rate consideration in wafer bonding”, under preparation.

Appendix

Verification of the Finite Element Method Simulation

In this appendix, an analytical plane strain solution of the elastic field in a completely bonded wafer with one-dimensional sinusoidal misfit gap was given. The finite element method (FEM) numerical results in Section 4.3 are compared with them. It is shown that our numerical simulation produced results accurate enough to be used in the discussion on the wafer bonding process.

Here, we consider the complete bonding between a wafer with one-dimensional sinusoidal surface and a rigid plane as shown in Fig. 4.3. The wafer thickness is t . Only elasticity is considered. The ratio Δ/λ is taken to be so small that the linear elasticity theory applies. Let (u_1, u_2, u_3) be the displacement field, which obeys Navier's equation:

$$(1 - 2\nu)u_{i,jj} + u_{j,ji} = 0 \quad (\text{A.1})$$

The stresses, σ_{ij} , relate to the displacements as

$$\sigma_{ij} = \frac{E}{1 + \nu} \left[\frac{1}{2} (u_{i,j} + u_{j,i}) + \frac{\nu}{1 - 2\nu} u_{k,k} \delta_{ij} \right] \quad (\text{A.2})$$

where δ_{ij} is Kronecker delta. $\delta_{ij} = 1$ when $i = j$, and $\delta_{ij} = 0$ when $i \neq j$.

As only plane strain problem in the xOz plane is considered, $u_2 = 0$.

To accommodate the sinusoidal misfit gap, we assume the displacement field in the wafer as

$$\begin{aligned} u_1 &= \sin(kx)f(kz) \\ u_3 &= w_0 + \cos(kx)g(kz) \end{aligned} \quad (\text{A.3})$$

where $k = 2\pi/\lambda$, and w_0 represents a rigid body translation. The functions f and g are determined by substituting Eq. (A.3) into the Navier's equation, giving

$$\begin{aligned}(1-2\nu)f''(kz) - 2(1-\nu)f(kz) - g'(kz) &= 0 \\ 2(1-\nu)g''(kz) - (1-2\nu)g(kz) + f'(kz) &= 0\end{aligned}\quad (\text{A.4})$$

They are ordinary differential equations of constant coefficients, to which the general solution is

$$\begin{aligned}f(kz) &= [C_1 + C_2 kz]e^{-kz} + [C_3 + C_4 kz]e^{kz} \\ g(kz) &= [C_1 + (3-4\nu+kz)C_2]e^{-kz} + [-C_3 + (3-4\nu-kz)C_4]e^{kz}\end{aligned}\quad (\text{A.5})$$

The coefficients C_i ($i = 1..4$) are to be determined by boundary conditions.

We apply the general solution to the wafer under the following boundary conditions: (i) on the free surface ($z = t$), the normal stress vanishes; (ii) on the free surface ($z = t$), the shear stress vanishes, (iii) on the interface ($z = 0$), the shear stress vanishes, (iv) on the interface ($z = 0$), the vertical displacement is

$$u_3 = -\Delta[1 - \cos(kx)] \quad (\text{A.6})$$

The four boundary conditions determines four coefficients, giving

$$\begin{aligned}C_1 &= -\frac{\Delta}{2(1-\nu)} \frac{(1-2\nu)(2kt-1+e^{2kt}) + 2k^2 t^2}{e^{2kt} + 4kt - e^{-2kt}} \\ C_2 &= -\frac{\Delta}{2(1-\nu)} \frac{1-2kt-e^{2kt}}{e^{2kt} + 4kt - e^{-2kt}} \\ C_3 &= \frac{\Delta}{2(1-\nu)} \frac{(1-2\nu)(2kt+1-e^{-2kt}) - 2k^2 t^2}{e^{2kt} + 4kt - e^{-2kt}} \\ C_4 &= -\frac{\Delta}{2(1-\nu)} \frac{e^{-2kt} - 2kt - 1}{e^{2kt} + 4kt - e^{-2kt}}\end{aligned}\quad (\text{A.7})$$

In the wafer, the stress component σ_{zz} is

$$\sigma_{zz} = -\frac{Ek\Delta \cos(kx)}{2(1-\nu^2)(e^{2kt} + 4kt - e^{-2kt})} \times \begin{bmatrix} (e^{2kt} + 2kt - 1 - 2k^2 t^2)e^{-kz} \\ + (e^{2kt} + 2kt - 1)kze^{-kz} \\ + (e^{-2kt} - 2kt - 1 - 2k^2 t^2)e^{kz} \\ + (1 + 2kt - e^{-2kt})kze^{kz} \end{bmatrix} \quad (\text{A.8})$$

On the interface ($z = 0$), it is

$$\sigma_{zz} = -\frac{Ek\Delta \cos(kx)}{2(1-\nu^2)} \times \frac{e^{2kt} + e^{-2kt} - 4k^2 t^2 - 2}{e^{2kt} + 4kt - e^{-2kt}} \quad (\text{A.9})$$

Its maximum is

$$(\sigma_{zz})_{\max} = \frac{Ek\Delta}{2(1-\nu^2)} \times \frac{e^{2kt} + e^{-2kt} - 4k^2 t^2 - 2}{e^{2kt} + 4kt - e^{-2kt}} \quad (\text{A.10})$$

Therefore, the stresses and displacements are directly related to the misfit gap amplitude, Δ .

For the limiting case where the wafer is so thick that $\lambda \ll t$, σ_{zz} is

$$\sigma_{zz} = -\frac{Ek\Delta \cos(kx)}{2(1-\nu^2)} (1 + kz)e^{-kz} \quad (\text{A.11})$$

which decays exponentially in the z direction. The normal stress on the interface is

$$\sigma_{zz} = -\frac{Ek\Delta \cos(kx)}{2(1-\nu^2)} \quad (\text{A.12})$$

Its maximum and minimum are:

$$(\sigma_{zz})_{\max} = \pm \frac{Ek\Delta}{2(1-\nu^2)} = \pm \frac{E\pi}{(1-\nu^2)} \frac{\Delta}{\lambda} \quad (\text{A.13})$$

Obviously they are linear in Δ/λ and the wafer elastic modulus.

The strain energy for complete bonding can be computed by the work done by σ_{zz} at the interface to close the misfit gap. For half a period of asperity, it is

$$U = \frac{1}{2} \int_0^l -u_3 \sigma_{zz} dx = \frac{E\pi\Delta^2}{8(1-\nu^2)} \times \frac{e^{2kt} + e^{-2kt} - 4k^2t^2 - 2}{e^{2kt} + 4kt - e^{-2kt}}$$

(A.14)

The maximum and minimum normal stresses on the interface and the strain energy per half period of asperity from our FEM simulation are very close to the corresponding analytical solutions, as shown in Table A.1.

Table A.1 Numerical and analytical results for plane strain complete bonding

($U_{\max}$: J/m; σ_{zz} : MPa)

$2\Delta/\lambda$	$U_{\max}$ (FEM)	$U_{\max}$ (analytical)	$(\sigma_{zz})_{\min}$ (FEM)	$(\sigma_{zz})_{\min}$ (analytical)	$(\sigma_{zz})_{\max}$ (FEM)	$(\sigma_{zz})_{\max}$ (analytical)
0.005	0.5393	0.5394	-432.1	-431.5	430.3	431.5
0.0025	0.1348	0.1349	-215.9	-215.8	215.4	215.8
0.0005	5.393×10-3	5.394×10-3	-43.13	-43.15	43.15	43.15

References

- [1] Talke, F.E. (1995), "On tribological problems in magnetic disk recording technology". *Wear*, Vol.190, pp.232-238.
- [2] Tai, Y.C. (1989), **IC-processed polysilicon micromechanics: technology, material, and devices**. PhD Dissertation, University of California.
- [3] Fan, L.S. (1990), **Integrated micromachinery --- moving structures on silicon chips**. PhD Dissertation, University of California.
- [4] Mehregany, M. (1990), **Microfabricated silicon electric mechanisms**. PhD Dissertation, Massachusetts Institute of Technology.
- [5] Orr, F.M., Scriven, L.E. and Rivas, A.P. (1975), "Pendular rings between solids: meniscus properties and capillary force". *Journal of Fluid mechanics*, Vol.67, part 4, pp.723-742.
- [6] Gösele, U., Tong, Q.-Y., Schumacher, A., Kräuter, G., Reiche, M., Plöchl, A., Kopperschmidt, P., Lee, T.-H. and Kim, W.-J. (1999), "Wafer bonding for microsystems technologies". *Sensors and Actuators A: Physical*, Vol.74, pp161-168.
- [7] Hui, C.Y., Baney, J.M. and Lin, Y.Y. (2001), "Mechanics of the JKR (Johnson-Kendall-Roberts) adhesion test". In: **Adhesion Measurement of Films and Coatings**, Vol.2, pp299-328. Ed. K.L. Mittal, ©VSP 2001.
- [8] Bhushan, B. (1996), "Contact mechanics of rough surfaces in tribology: Single asperity contact". *Applied Mechanics Review*, Vol.49, No.5, pp.275-298.
- [9] Bhushan, B. (1996), **Tribology and Mechanics of Magnetic Storage Devices**, 2nd Ed. New York: Springer.
- [10] Bhushan, B. (1996), "Methodology for roughness measurement and contact analysis for optimization of interface roughness". *IEEE Transactions on Magnetics*, Vol.32, No.3, Pt.2, pp.1819-1825.

-
- [11] Bhushan, B. (1998), "Contact mechanics of rough surfaces in tribology: multiple asperity contact". *Tribology Letters*, Vol.4, No.1, pp.1-35.
 - [12] Bhushan, B. (1999), **Principles and applications of tribology**, New York, USA: John Wiley & Sons.
 - [13] Bhushan, B. (2001), "Tribology on the macroscale to nanoscale of microelectromechanical system materials: A review". *Proceedings of the Institution of Mechanical Engineers, Part J: Journal of Engineering Tribology*, Vol.215, No.1, pp.1-18.
 - [14] Gao, C., Dai, P., Homola, A., and Weiss, J. (1998), "Meniscus force and profiles: theory and its applications to liquid-mediated interfaces". *Journal of Tribology — Transactions of the ASME*, Vol.120, pp.358-368.
 - [15] Derjaguin, B.V., Churaev, N.V., and Muller, V.M. (1987), **Surface Forces**. New York: Consultants Bureau.
 - [16] Israelachvili, J.N. (1992), **Intermolecular and Surface Forces**, 2nd Ed. San Diego, CA: Academic Press.
 - [17] Haisma, J., Spierings, B.A.M., Biermann, U.K.P. and van Gorkum, A.A. (1994), "Diversity and feasibility of direct bonding: a survey of a dedicated optical technology". *Applied Optics*, Vol.33, pp1154-1169.
 - [18] Babic, D.I., Dudley, J.J., Streubel, K., Mirin, R.P., Bowers, J.E. and Hu, E.L. (1995), "Double-fused 1.52 μm vertical-cavity lasers". *Applied Physics Letters*, Vol.66, pp1030-1032.
 - [19] Liao, Z.L. (1997), "Strained interface of lattice-mismatched wafer fusion". *Physical Review B: Condensed Matter and Materials Physics*, Vol.55, No.19, pp.12899–12901.
 - [20] Gösele, U., Stenzel, H., Martini, T., Steinkirchner, J., Conrad, D. and Scheerschmidt, K. (1995), "Self-propagating room-temperature silicon wafer bonding in ultrahigh vacuum". *Applied Physics Letters*, Vol.67, Issue 24, pp3614-3616.

-
- [21] Tong, Q.-Y. and Gösele, U. (1994), "Semiconductor wafer bonding: recent developments". *Materials Chemistry and Physics*, Vol.37, No.2, pp.101-127.
 - [22] Gui, C., Elwenspoek, M., Tas, N. and Gardeniers, J.G.E. (1999), "The effect of surface roughness on direct wafer bonding". *Journal of Applied Physics*, Vol.85, No.10, pp7448-7454.
 - [23] Wada, H., Ogawa, Y. and Kamijoh, T. (1993), "Electrical characteristics of directly-bonded GaAs and InP". *Applied Physics Letters*, Vol.62, pp738-740.
 - [24] Bowden, F.P. and Tabor, D. (1954), **The friction and lubrication of solids, Part I**, Oxford: Clarendon Press.
 - [25] Finn, R. (1986), **Equilibrium Capillary Surfaces**. New York: Springer-Verlag.
 - [26] Lian, G.P., Thorton, C., and Adams, M.J. (1993), "A theoretical study of the liquid bridge forces between two rigid spherical bodies". *Journal of Colloid and Interface Science*, Vol.161, Issue 1, pp.138-147.
 - [27] Gao, C., Tian, X. and Bhushan, B. (1995), "A meniscus model for optimization of texturing and liquid lubrication of magnetic thin film rigid disks". *Tribology Transactions*, Vol.38, pp.201-212.
 - [28] Tian, X. and Bhushan, B. (1996), "A numerical three-dimensional model for the contact of rough surfaces by variational principle". *Journal of Tribology — Transactions of the ASME*, Vol.118, pp.33-42.
 - [29] Tian, X. and Bhushan, B. (1996), "The micro-meniscus effect of a thin liquid film on the static friction of rough surface contact". *Journal of Physics D: Applied Physics*, Vol.29, pp.163-178.
 - [30] Stengl, R., Mitani, K., Lehmann, V. and Gösele, U. (1989), "Silicon Wafer Bonding: Chemistry, Elastomechanics and Manufacturing". In: **Proceedings of IEEE SOS/SOI technology conference**, Stateline, Nevada, pp123-124.
 - [31] Gösele, U. and Tong, Q.-Y. (1993), "Silicon layer transfer by wafer bonding". In: **Proceedings of the 2nd International Symposium on Semiconductor Wafer Bonding: Science, Technology, and Applications**, edited by M. A.

-
- Schmidt, T. Abe, C. E. Hunt, and H. Baumgart, PV 93-29, (The Electrochemical Society Proceedings Series, Pennington, NJ, 1993), pp395-409.
- [32] Tong, Q.-Y. and Gösele, U. (1995), "Thickness considerations in direct silicon wafer bonding". *Journal of the Electrochemical Society*, Vol.142, No.11, pp.3975-3979.
- [33] Yu, H.H. and Suo, Z. (1998), "A model of wafer bonding by elastic accommodation". *Journal of the Mechanics and Physics of Solids*, Vol.46, No.5, pp829-844.
- [34] Johnson, K.L. (1985), **Contact mechanics**, Cambridge, UK: Cambridge University Press.
- [35] Johnson, K.L., Kendall, K. and Roberts, A.D. (1971), "Surface energy and the contact of elastic solids". *Proceedings of the Royal Society of London, Series A, Mathematical and Physical Sciences*, Vol.A324, pp301-313.
- [36] Suo, Z. (1997), "Motions of microscopic surfaces in materials". *Advances in Applied Mechanics*, Vol.33, pp.194-289.
- [37] Poon, C.Y. and Bhushan, B. (1996), "Numerical contact and stiction analysis of Gaussian isotropic surfaces for magnetic head slider/disk contact". *Wear*, Vol.202, pp.68-82.
- [38] Fan, H. and Gao, Y.X. (2001), "Elastic solution for liquid-bridging-induced microscale contact". *Journal of Applied Physics*, Vol.90, No.12, pp.5904-5910.
- [39] Yang, W. and Suo, Z. (1996), "Global view of microstructural evolution: energetics, kinetics and dynamical systems". *Acta Mechanica Sinica (English Series)*, Vol.12, No.2, pp.144-157.
- [40] Sun, B., Suo, Z., and Cocks, A.C.F. (1996), "A global analysis of structural evolution in a row of grains". *Journal of the Mechanics and Physics of Solids*, Vol.44, No.4, pp.559-581.
- [41] Loge, R.E. and Suo, Z. (1996), "Nonequilibrium thermodynamics of ferroelectric domain evolution". *Acta Materialia*, Vol.44, No.8, pp.3429-3438.

-
- [42] Herring, C. (1951), "Surface tension as a motivation for sintering". In: **The Physics of Powder Metallurgy**, Ed. W.E. Kingston. New York: McGraw-Hill Book Co.
 - [43] Bhushan, B. and Zhao, Z. (1997), "Friction/stiction and wear studies of magnetic thin-film disks with two perfluoropolyether lubricants". *IEEE Transactions on Magnetics*, Vol.31, No.1, pp.918–925.
 - [44] Bhushan, B., Kotwal, C.A. and Chilamakuri, S.K. (1998), "Kinetic meniscus model for prediction of rest stiction". *ASME Journal of Tribology*, Vol.120, No.1, pp.42-50.
 - [45] **ANSYS User's manuals for Release 5.5**, (1998), Canonsburg, USA: ANSYS Inc.
 - [46] **Origin User's manual for Version 6.1** (2000), Northampton, USA: OriginLab Corporation.
 - [47] Deryagin, B.V., Muller, V.M. and Toporov, Yu.P. (1975), "Effect of contact deformations on the adhesion of particles". *Journal of Colloid and Interface Science*, Vol.53, pp314-326.
 - [48] Maugis, D. (1992), "Adhesion of spheres: the JKR-DMT transition using a Dugdale model". *Journal of Colloid and Interface Science*, Vol.150, No.1, pp.243-269.
 - [49] Maszara, W.P., Goetz, G., Caviglia, A. and McKitterick, J.B. (1988), "Bonding of silicon wafers for silicon-on-insulator". *Journal of Applied Physics*, Vol.64, pp.4943–4950.
 - [50] Griffith, A.A. (1921), "The phenomena of rupture and flow in solids". *Philosophical Transactions of the Royal Society of London*, Vol.A221, pp163-198.
 - [51] Fan, H. and Sze, K.Y. (2001), "A micro-mechanics model for imperfect interface in dielectric materials". *Mechanics of Materials*, Vol.33, No.6, pp.363-370.

-
- [52] Adamson, A.W. (1990), **Physical Chemistry of Surfaces**. New York: J. Wiley.
 - [53] Barber, J.R. and Ciavarella, M. (2000), "Contact mechanics". *International Journal of Solids and Structures*, Vol.37, pp29-43.
 - [54] Adams, G.G. and Nosonovsky, M. (2000), "Contact modeling — forces". *Tribology International*, Vol.33, pp.431-442.
 - [55] Ando, Y. (2000), "The effect of relative humidity on friction and pull-off forces measured on submicron-size asperity arrays". *Wear*, Vol.238, pp.12-19.
 - [56] Ando, Y. and Ino, J. (1997), "The effect of asperity array geometry on friction and pull-off force". *Journal of Tribology — Transactions of the ASME*, Vol.119, No.4, pp.781-787.
 - [57] Baets, P.D., Degrieck, J., Velde, F.V.D. and Peteghem, A.P.V. (2000), "Experimental verification of the mechanisms causing stick-slip motion originating from relative deceleration". *Wear*, Vol.243, pp.48-59.
 - [58] Bai, M.W. and Kato, K. (2001), "Analysis of contact deformation and stiction between textured disk and textured slider". *Journal of Tribology — Transactions of the ASME*, Vol.123, No.2, pp.350-357.
 - [59] Bailey, D.M and Sayles, R.S. (1991), "Effect of roughness and sliding friction on contact stresses". *Journal of Tribology — Transactions of the ASME*, Vol.113, pp.729-738.
 - [60] Bjorklund, S.A. (1997), "A random model for micro-slip between nominally flat surfaces". *ASME Journal of Tribology*, Vol.119, pp.726-32.
 - [61] Blencoe, K.A., Roper, G.W. and Williams, J.A. (1998), "The influence of lubricant rheology and surface topography in modeling friction at concentrated contacts". *Proceedings of the Institution of Mechanical Engineers, Part J: Journal of Engineering Tribology*, Vol.212, pp.391-400.
 - [62] Busquet, M. and Torrance, A. (2000), "A numerical slipline field for the sliding cylinder problem". *Wear*, Vol.241, pp.86-98.

-
- [63] Decenciere, E. and Jeulin, D. (2001), "Morphological decomposition of the surface topography of an internal combustion engine cylinder to characterize wear". *Wear*, Vol.249, pp.482-488.
 - [64] Faulkner, A. and Arnell, R.D. (2000), "The development of a finite element model to simulate the sliding interaction between two, three-dimensional, elastoplastic, hemispherical asperities". *Wear*, Vol.242, pp.114-122.
 - [65] Fujimoto, T., Kagami, J., Kawaguchi, T. and Hatazawa, T. (2000), "Micro-displacement characteristics under tangential force". *Wear*, Vol.241, pp.136-142.
 - [66] Green, A.P. (1954), "Friction between unlubricated metals: a theoretical analysis of the junction model".
 - [67] Greenwood, J.A. (1997), "Analysis of elliptical Hertzian contacts". *Tribology International*, Vol.30, pp.235-237.
 - [68] Hagman, L.A. and Olofsson, U. (1998), "A model for micro-slip between flat surfaces based on deformation of elliptical elastic asperities — parametric study and experiment investigation". *Tribology International*, Vol.31, pp.209-217.
 - [69] Handzel-Powierza, Z., Klimczak, T. and Polijaniuk, A. (1992), "On the experimental verification of the Greenwood–Williamson model for the contact of rough surfaces". *Wear*, Vol.154, pp.115–124.
 - [70] Horng, J.H. (1998), "An elliptic elastic-plastic asperity model for rough surfaces". *Journal of Tribology — Transactions of the ASME*, Vol.120, pp.82-88.
 - [71] Kral, E.R., Komvopoulos, K., and Body, D.B. (1995), "Finite element analysis of repeated indentation of an elastic-plastic layered medium by a rigid sphere, Part I: Surface results". *Journal of Applied Mechanics — Transactions of the ASME*, Vol.62, pp.20-28.
 - [72] Kral, E.R., Komvopoulos, K., and Body, D.B. (1995), "Finite element analysis of repeated indentation of an elastic-plastic layered medium by a rigid sphere,

- Part II: Substrate results”. *Journal of Applied Mechanics — Transactions of the ASME*, Vol.62, pp.29-42.
- [73] Kotwal, C.A. and Bhushan, B. (1996), “Contact analysis of non-Gaussian surfaces for minimum static and kinetic friction and wear”. *Tribology Transactions*, Vol.39, pp.890-898.
- [74] Le, H.R. and Sutcliffe, M.P.F. (2000), “Analysis of surface roughness of cold-rolled aluminum foil”. *Wear*, Vol.244, pp.71-78.
- [75] Liu, Z.Q., Neville, A. and Reuben, R.L. (2001), “A numerical calculation of the contact area and pressure of real surfaces in sliding wear”. *Journal of Applied Mechanics — Transactions of the ASME*, Vol.123, pp.27-35.
- [76] Lu, X.C., Shi, B., Li, L.K.Y., Luo, J.B. and Mou, J.I. (2001), “Nanoindentation and nanotribological behavior of Fe-N/Ti-N multilayers with different thickness of Fe-N layers”. *Wear*, Vol.247, pp.15-23.
- [77] Majumdar, A. and Bhushan, B. (1991), “Fractal model of elastic-plastic contact between rough surfaces”. *Journal of Tribology — Transactions of the ASME*, Vol.113, pp.1-11.
- [78] Manners, W. (1998), “Partial contact between elastic surfaces with periodic profiles”. *Proceedings of the Royal Society of London, Series A (Mathematical, Physical and Engineering Sciences)*, Vol.454, pp.3203-3221.
- [79] Mokhtar, M.O.A., Younes, Y.K., Mahdy, T.H.E. and Attia, N.A. (1998), “A theoretical and experimental study on the dynamics of sliding bodies with dry conformal contacts”. *Wear*, Vol.218, pp.172-178.
- [80] Morrow, C., Lovell, M. and Zhi, D. (2001), “Numerical contact analysis of transversely isotropic coatings: a cylinder within a circumferential groove”. *Journal of Tribology — Transactions of the ASME*, Vol.123, pp.436-440.
- [81] Nosonovsky, M. and Adams, G.G. (2000), “Steady state frictional sliding of two elastic bodies with a wavy contact surface”. *Journal of Tribology — Transactions of the ASME*, Vol.122, pp.490-495.

-
- [82] O'Sullivan, T.C. and King, R.B. (1988), "Sliding contact stress field due to a spherical indenter on a layered elastic half-space". *Journal of Tribology — Transactions of the ASME*, Vol.110, pp.235-240.
 - [83] Othmani, A. and Kaminsky, C. (1998), "Three dimensional fractal analysis of sheet metal surface". *Wear*, Vol.214, pp.147-150.
 - [84] Podsiadlo, P. and Stachowiak, G.W. (1998), "Evaluation of boundary fractal methods for the characterization of wear particles". *Wear*, Vol.217, 24-34.
 - [85] Poon, C.Y. and Bhushan, B. (1996), "Nano-asperity contact analysis and surface optimization for magnetic head slider/disk contact". *Wear*, Vol.202, pp.83-98.
 - [86] Robbe-Valloire, F. (2001), "Statistical analysis of asperities on a rough surface". *Wear*, Vol.249, pp.401-408.
 - [87] Roques-Carmes, C., Bodin, N., Monteil, G. and Quiniou, J.F. (2001), "Description of rough surfaces using conformal equivalent structure concept Part 1. Stereological approach". *Wear*, Vol.248, pp.82-91.
 - [88] Roques-Carmes, C., Bodin, N., Monteil, G. and Quiniou, J.F. (2001), "Description of rough surfaces using conformal equivalent structure concept Part 2. Numerical approach". *Wear*, Vol.248, pp.92-99.
 - [89] Turner, K.T. and Spearing, S.M. (2002), "Modeling of direct wafer bonding: Effect of wafer bow and etch patterns". *Journal of Applied Physics*, Vol.92, No.12, pp7658-7666.
 - [90] Tworzydło, W.W., Cecot, W., Oden, J.T. and Yew, C.H. (1998), "Computational micro- and macroscopic models of contact and friction: formulation, approach and applications". *Wear*, Vol.220, pp.113-140.
 - [91] Peng, W. and Bhushan, B. (2001), "A numerical three-dimensional model for the contact of layered elastic/plastic solids with rough surfaces by a variational principle". *Journal of Tribology — Transactions of the ASME*, Vol.123, pp.330-342.

-
- [92] Zhao, Y., Maietta, D.M. and Chang, L. (2000), "An asperity microcontact model incorporating the transition from elastic deformation to fully plastic flow". *Journal of Tribology — Transactions of the ASME*, Vol.122, pp.86-93.
 - [93] Rymuza, Z. (1999), "Control tribological and mechanical properties of MEMS surfaces. Part 1: Critical review". *Microsystem Technologies*, Vol.5, No.4, pp.173-180.
 - [94] Rymuza, Z., Misiak, M., Kuhn, L., Schmidt-Szalowski, K., Rzanek-Boroch, Z. (1999), "Control tribological and mechanical properties of MEMS surfaces. Part 2: Nanomechanical behavior of self-lubricating ultrathin films". *Microsystem Technologies*, Vol.5, No.4, pp.181-188.
 - [95] Decencire, E. and Jeulin, D. (2001), "Morphological decomposition of the surface topography of an internal combustion engine cylinder to characterize wear". *Wear*, Vol.249, pp.482-488.
 - [96] Tan, S. and Talke, F.E. (2001), "Numerical and experimental investigations of the head/tape interface in a digital linear tape drive". *Journal of Tribology — Transactions of the ASME*, Vol.123, No.2, pp.343-349.
 - [97] Dwyer-Joyce, R.S., Drinkwater, B.W. and Quinn, A.M. (2001), "The use of ultrasound in the investigation of rough surface interfaces". *Journal of Tribology — Transactions of the ASME*, Vol.123, No.1, pp.8-16.
 - [98] Liu, S. and Wang, Q. (2001), "A three-dimensional thermomechanical model of contact between non-conforming rough surfaces". *Journal of Tribology — Transactions of the ASME*, Vol.123, No.1, pp.17-26.
 - [99] Yang, P., Qu, S., Chang, Q. and Guo, F. (2001), "On the theory of thermal elastohydrodynamic lubrication at high slide-roll ratios - Line contact solution". *Journal of Tribology — Transactions of the ASME*, Vol.123, No.1, pp.36-41.
 - [100] Liu, Y. and German, R.M. (1996), "Contact angle and solid-liquid-vapor equilibrium". *Acta Materialia*, Vol.44, No.4, pp.1657-1663.
 - [101] Bourago, N.G. (2002), "A Survey on Contact Algorithms". In: **Proceedings of Workshop "Grid Generation: Theory and Applications"**, Edited by S.A.

-
- Ivanenko & V.A.Garanzha, Computing Centre of RAS, Moscow, July, 2002, pp.42-59.
- [102] Kogut, L. and Etsion, I. (2002), "Elastic-Plastic Contact Analysis of a Sphere and a Rigid Flat". *Journal of Applied Mechanics*, Vol.69, pp.657-662.
- [103] Kogut, L. and Etsion, I. (2002), "An improved elastic-plastic model for the contact of rough surfaces". Presented in: **3rd AIMETA international tribology conference**, VIETRI SUL MARE, SALERNO, Italy, 18-20 September, 2002.
- [104] Peng, W. and Bhushan, B. (2002), "Sliding contact analysis of layered elastic/plastic solids with rough surfaces". *Journal of Tribology — Transactions of the ASME*, Vol.124, No.1, pp.46-61.
- [105] Kogut, L. and Etsion, I. (2003), "A semi-analytical solution for the sliding inception of a spherical contact". *Journal of Tribology — Transactions of the ASME*, Vol.125, No.3, pp.499-506.
- [106] Zou, P., Scherge, M. and Lambeth, D.N. (1995), "Quasi-contact testing of carbon nitride hard disk overcoats". *IEEE Transactions on Magnetics*, Vol.31, No.6, Pt.1, pp.2985-2987.
- [107] Pimenov, S.M., Smolin, A.A., Obraztsova, E.D., Konov, V.I., Bögli, U., Blatter, A., Loubnin, E.N., Maillat, M., Leijala, A., Burger, J. and Hintermann, H.E. (1996), "Tribological properties of smooth diamond films". *Applied Surface Science*, In: **Proceedings of the Seventh International Conference on Solid Films and Surfaces**, Edited by H.L. Hwang and T.T. Tsong, Hsinchu, Taiwan, ROC, 12 - 16 December 1994, pp.106-114.
- [108] Shull, K.R., Ahn, D. and Mowery, C.L. (1997), "Finite-Size Corrections to the JKR Technique for Measuring Adhesion: Soft Spherical Caps Adhering to Flat, Rigid Surfaces". *Langmuir*, Vol.13, pp.1799-1804.
- [109] Sridhar, I., Johnson, K.L. and Fleck, N.A. (1997), "Adhesion mechanics of the surface force apparatus". *Journal of Physics D: Applied Physics*, Vol.30, No.12, pp.1710-1719.

-
- [110] Johnson, K.L. and Sridhar, I. (2001), "Adhesion between a spherical indenter and an elastic solid with a compliant elastic coating". *Journal of Physics D: Applied Physics*, Vol.34, No.5, pp.683-689.
 - [111] Gao, Y.X., Fan, H. and Xiao, Z. (2000), "A thermodynamics model for solder profile evolution". *Acta Materialia*, Vol.48, No.4, pp.863-874.
 - [112] Huang, P., Li, Z. and Sun, J. (2001), "Finite element analysis for surface diffusion-controlled shape instabilities of plate-like grains". *Computational Materials Science*, Vol.20, No.1, pp.66-76.
 - [113] Wan, K.-T. and Liu, K.-K. (2001), "Contact mechanics of a thin-walled capsule adhered onto a rigid planar substrate". *Medical & Biological Engineering & Computing*, Vol.39, No.5, pp.605-608.
 - [114] Liu, K.K. , Wang, H.G., Wan, K.T., Liu, T. and Zhang, Z. (2002), "Characterizing capsule-substrate adhesion in presence of osmosis". *Colloids and Surfaces B: Biointerfaces*, Vol.25, No.4, pp.293-298.
 - [115] Gao, Y.X. and Fan, H. (2002), "A micro-mechanism based analysis for size-dependent indentation hardness". *Journal of Materials Science*, Vol.37, No.20, pp. 4493-4498.
 - [116] Gao, Y.F., Lu, W. and Suo, Z. (2002), "A mesophase transition in a binary monolayer on a solid surface". *Acta Materialia*, Vol.50, pp. 2297–2308.
 - [117] Gao, Y.X., Fan, H. and Xiao, Z. (2000), "A simulation of asymmetrical voids evolution induced by electromigration". *Mechanics of Materials*, Vol.32, pp.315-326.
 - [118] Kralchevsky, P.A. and Denkov, N.D. (2001), "Capillary forces and structuring in layers of colloid particles". *Current Opinion in Colloid & Interface Science*, Vol.6, No.4, pp.383-401.
 - [119] Etsion, I. (2000), "Discussion: "An Asperity Microcontact Model Incorporating the Transition from Elastic Deformation to Fully Plastic Flow"". *Journal of Tribology — Transactions of the ASME*, Vol.122, No. 1, pp.86-93.

-
- [120] Zhao, Y.W., Marietta, D.M. and Chang, L. (2000), "Closure to "discussion of an asperity microcontact model incorporating the transition from elastic deformation to fully plastic flow"". *Journal of Tribology — Transactions of the ASME*, Vol.122, No.2, pp.479-480.
 - [121] Hillert, M. and Agren, J. (2002), "Effect of surface free energy and surface stress on phase equilibria". *Acta Materialia*, Vol.50, No.9, pp.2429-2441.
 - [122] Kaneko, S., Ikeda, T., Saito, T. and Ito, S. (2003), "Experimental study on static and dynamic characteristics of liquid annular convergent-tapered damper seals with honeycomb roughness pattern". *Journal of Tribology — Transactions of the ASME*, Vol.125, No.3, pp.592-599.
 - [123] Saiz, E., Tomsia, A.P. and Cannon, R.M. (1998), "Ridging effects on wetting and spreading of liquids on solids". *Acta Materialia*, Vol.46, No.7, pp.2349-2361.
 - [124] Podsiadlo, P. and Stachowiak, G.W. (2000), "Scale-invariant analysis of wear particle surface morphology. I: Theoretical background, computer implementation and technique testing". *Wear*, Vol.242, No.1, pp.160-179.
 - [125] Podsiadlo, P. and Stachowiak, G.W. (2000), "Scale-invariant analysis of wear particle surface morphology. II: Fractal dimension". *Wear*, Vol.242, No.1, pp.180-188.
 - [126] Podsiadlo, P. and Stachowiak, G.W. (2000), "Scale-invariant analysis of wear particle surface morphology. III: Pattern recognition". *Wear*, Vol.242, No.1, pp.189-201.
 - [127] Cule, D. and Hwa, T. (1996), "Tribology of sliding elastic media". *Physical Review Letters*, Vol.77, No.2, pp.278-281.
 - [128] Xia, L., Bower, A.F., Suo, Z. and Shih, C.F. (1997), "A finite element analysis of the motion and evolution of voids due to strain and electromigration induced surface diffusion". *Journal of the Mechanics and Physics of Solids*, Vol.45, No.9, pp.1473-1493.

-
- [129] Sun, B., Suo, Z. and Yang, W. (1997), "A finite element method for simulating interface motion. I: Migration of phase and grain boundaries". *Acta Materialia*, Vol.45, No.5, pp.1907-1915.
 - [130] Sun, B. and Suo, Z. (1997), "A finite element method for simulating interface motion. II: Large shape change due to surface diffusion". *Acta Materialia*, Vol.45, No.12, pp.4953-4962.
 - [131] Parhami, F., McMeeking, R.M., Cocks, A.C.F. and Suo, Z. (1999), "A model for the sintering and coarsening of rows of spherical particles". *Mechanics of Materials*, Vol.31, No.1, pp.43-61.
 - [132] Suo, Z. and Yu, H. (1997), "Crack nucleation on an elastic polycrystal surface in a corrosive environment: low dimensional dynamical models". *Acta Materialia*, Vol.45, No.6, pp.2235-2245.
 - [133] Suo, Z. and Wang, W. (1994), "Diffusive void bifurcation in stressed solid". *Journal of Applied Physics*, Vol.76, No.6, pp.3410-3421.
 - [134] Wang, Weiqing and Suo, Z. (1997), "Shape change of a pore in a stressed solid via surface diffusion motivated by surface and elastic energy variation". *Journal of the Mechanics and Physics of Solids*, Vol.45, No.5, pp.709-729.
 - [135] Suo, Z. (1989), "Singularities, interfaces and cracks in dissimilar anisotropic media". *Proceedings of the Royal Society of London, Series A (Mathematical and Physical Sciences)*, Vol.427, No.1873, pp.331-358.
 - [136] Fan, H., Gao, Y.X. and Huang, X.Y. (2001), "Thermodynamics modeling for moving contact line in gas/liquid/solid system: Capillary rise problem revisited". *Physics of Fluids*, Vol.13, No.6, pp.1615-1623.