

Quantitative Analysis and Predictive Engineering of

Self-rolling of Strain Engineered Nanomembranes

by

Cheng Chen

Department of Mining and Materials Engineering McGill University, Montréal

April 2018

A thesis submitted to McGill University in partial fulfillment of the requirements of the degree of Doctor of Philosophy

© Cheng Chen, 2018

Abstract

With excellent geometrical flexibility and versatility applicable to various functional materials, self-rolling of strain engineered nanomembranes promises unprecedented possibilities for creating complicated three-dimensional (3D) geometries through programmable shape transformations and attracted enormous scientific interests. It perfectly combines top-down and bottom-up methods, enabling diverse technological applications ranging from nano-elecromechanical/micro-electromechanical systems (NEMS/MEMS) and actuators/sensors, to microrobotics and biotechnology. Challenges remains as understanding of the mechanisms underlying the self-rolling process remains inadequate, which makes it difficult to control geometries of the resultant nanostructures and integrate state-of-the-art functionality required by those applications, thus rendering predictive design strategies of nanostructures from self-rolling not viable.

Therefore, aiming to have a deeper and more precise understanding the self-rolling behaviors of strain engineered nanomembranes towards accurately designing complex 3D structures, the present thesis systematically investigated various key aspects, including anisotropic mismatch strain, misfit dislocations, predefined surface patterns, and compositional inhomogeneity affecting the self-rolling process of strain engineered nanomembrane. Combing continuum modeling, numerical and atomistic simulations, the underlying mechanics and physics controlling the self-rolling process were systematically investigated, and predictive mechanics models were developed to provide a generic theoretical framework to guide the design of rolled-up nanostructures.

The thesis is manuscript-based, including the investigation of dependence of self-rolling behaviors of nanomembranes on anisotropic mismatch strain (Chapter 4), surface patterning induced curvature variation and rolling direction selection of self-rollup configurations (Chapter 5), misfit dislocation induced mismatch strain relaxation effect (Chapter 6), and effect of compositional inhomogeneity on self-rolling of nanomembranes (Chapter 7). The results of the thesis provide new mechanistic insights towards understanding self-rolling of strain engineered nanomembranes, providing critical guidance for the design and optimization of novel 3D nanomembrane structures.

Résumé

Avec une excellente flexibilité géométrique et une polyvalence le rendant applicable à divers matériaux fonctionnels, l'auto-enroulement de nano-membranes à base de contraintes promet des possibilités sans précédents pour créer des géométries tridimensionnelles (3D) compliquées par la programmation de changements de forme et il a attiré d'énormes intérêts scientifiques. Il combine parfaitement des méthodes descendantes et ascendantes, en permettant des applications technologiques diverses allant des systèmes nano-électromécaniques/micro-électromécaniques (NEMS/MEMS) aux actionneurs/capteurs, en passant par la microrobotique et les biotechnologies. Cependant, des défis persistent car la compréhension des mécanismes sous-jacents reste inadéquate dans le processus d'auto-roulant, ce qui rend difficile le contrôle des géométries des nanostructures qui en résultent et ce qui complique l'intégration de fonctionnalités de pointe requises pour ces applications, rendant ainsi les stratégies de conception prédictives des nanostructures auto-roulant non viables.

Ainsi, afin d'avoir une compréhension plus profonde et plus précise des comportements d'auto-enroulement des nanomembranes à base de contraintes pour la conception précise de structures 3D complexes, la présente thèse étudie systématiquement différents aspects clés, y compris la tension de mésappariement anisotrope, les dislocations inadaptées, les modèles de surface prédéfinis et l'inhomogénéité de la composition affectant le processus d'autoenroulement des nanomembranes. Combinant la modélisation en milieu continu, ainsi que les simulations numériques et atomistiques, la mécanique et la physique sous-jacentes qui contrôlent le processus auto-roulant ont été systématiquement étudiées et des modèles de mécanique prédictive ont été développés pour fournir un cadre théorique générique pour guider la conception des nanostructures. La thèse est fondée sur des manuscrits, qui incluent l'étude de la dépendance des comportements auto-enroulant des nano-membranes à l'égard de la tension de mésappariement anisotrope (Chapitre 4), de la configuration de surface induite par la variation de courbure et la sélection de la direction de laminage des configurations auto-rollup (chapitre 5), des dislocations inadaptées qui induisent un effet de relaxation des contraintes de mésappariement (Chapitre 6) et de l'inhomogénéité de la composition (Chapitre 7). Les résultats de la thèse apportent de nouvelles connaissances mécanistiques sur la compréhension du roulage automatique des nanomembranes à base de contraintes, fournissant des conseils critiques pour la conception et l'optimisation de nouvelles structures de nanomembranes 3D.

Acknowledgements

This thesis would not have been possible without the guidance and encouragement of my supervisor Prof. Jun Song, who is a person with true wisdom. The expertise, passion and patience he shows about scientific research have kept me motivated to continuous improvement in academic performance. It is very enjoyable for working with him in this distinguished research group.

I must also acknowledge my important coauthors, Prof. Xinyu Liu and Dr. Pengfei Song at University of Toronto, and group members Dr. Fanchao Meng and Mr. Pengfei Ou for their contributions to my work.

I appreciate the financial Assistance from China Scholarship Council (CSC) and McGill Engineering Doctoral Award. I also thank Supercomputer Consortium Laval UQAM McGill and Eastern Quebec (CLUMEQ) for providing computing power for all the simulation work in this thesis.

I would like to express my most special gratitude to my parents, Mr. Fuhua Chen and Mrs. Jinping Chen. Their unparalleled love and support made me concentrate on my research without being distracted.

Contributions of Authors

The thesis is manuscript-based, consisting of four coherent articles of the author's original work, which together with the contributions of all the authors are listed below (* indicates the corresponding author):

1. Quantitative analysis and predictive engineering of self-rolling of nanomembranes under anisotropic mismatch strain, *Nanotechnology*, 2017, 28 (48), 485302.

By: Cheng Chen, Pengfei Song, Fanchao Meng, Xiao Li, Xinyu Liu, Jun Song*

 Effect of Topological Patterning on Self-Rolling of Nanomembranes, Nanotechnology 29 (2018), 345301.

By: Cheng Chen, Pengfei Song, Fanchao Meng, Pengfei Ou, Xinyu Liu, Jun Song*

3. Misfit dislocation induced strain relaxation effect on self-rolling of strain-engineered nanomembranes, to be submitted.

By: Cheng Chen, Pengfei Song, Fanchao Meng, Pengfei Ou, Xinyu Liu, Jun Song*

4. Effects of inhomogeneity on the self-rolling of strained bilayer membranes, to be submitted.

By: Cheng Chen, Pengfei Song, Fanchao Meng, Pengfei Ou, Xinyu Liu, Jun Song*

Author contributions: For the above four papers, Cheng Chen and Jun Song conceived the ideas; Cheng Chen performed all the atomic simulations and continuum modeling under the supervision of Jun Song; Cheng Chen wrote the manuscript and Jun Song edited the text. Fanchao Meng assisted in the discussion of the setup of atomistic and continuum models. Xinyu Liu and Pengfei Song conducted related experiments to complement and support to the theoretical analysis. In Paper #1, Xiao Li assisted in the design of microfabrication experiments of rolled-up nanomembranes. In paper 2-4, Pengfei Ou contributed in the analysis and discussion of the results. Two additional journal articles had been published and one manuscript is in preparation, as a result of my Ph.D. research. They are related to this thesis but not included as the main chapters. Information regarding those articles is given below:

• Effects of Mg and Al Doping on Dislocation Slips on GaN, *Journal of Applied Physics*, 119, 064302:1-8, Cheng Chen, Fanchao Meng, and Jun Song*

• Core Structures Analysis of (a+c)-Edge Dislocations in Wurtzite GaN through Atomistic Simulations and Peierls-Nabarro Model, *Journal of Applied Physics*, 117, 194301:1-7, Cheng Chen, Fanchao Meng, and Jun Song*

• Rolled-up SiO_x/SiN_x microtube made from plasma enhanced chemical vapor deposition (PECVD) for sensitive solvent sensing, (accepted for publication of *Nanotechnology*), Pengfei Song, Cheng Chen, Juntian Qu, Pengfei Ou, Hadi Tavakoli Dastjerd, Zetian Mi, Jun Song, and Xinyu Liu

Table of Contents

Abst	ract		i				
Résu	mé		iii				
Ackr	nowled	gements	v				
Cont	ributio	ons of Authors	vi				
Tabl	e of Co	ontents	viii				
List	of Figu	res	xi				
List	of Tab	les	XX				
Chap	oter 1:	Introduction	1				
1.1	Refe	rences	4				
Chap	pter 2:	Literature Review	6				
2.1	Rolle	ed-up nanotechnology	6				
2.2	2.2 Generic design method of self-rolled-up structures						
2	2.2.1	Mechanisms of strain induced self-rolling of nanomembranes					
2.2.2		Rolling direction competition for rectangular nanomembranes					
2	2.2.3	Controllable curvature of self-rolled-up structures	13				
2	2.2.4	Computational modeling of self-rolled-up structures	18				
2.3	Prac	tical applications of strain induced self-rolled-up structures					
2	2.3.1	Self-rolled-up microtube used for microcavity resonators	21				
2	2.3.2	Compact on-chip self-rolled-up microtubes for lab-in-a-tube systems					
2.4 Self-		rolled-up nanomembranes used for energy storage devices	25				
2	2.4.1	Rolled-up nanomembranes used for magnetic tubular architectures	27				
2	2.4.2	Self-rolling method extended for the application of polymer structures	29				
2.5	Refe	rences	32				
Chap	pter 3:	Research Methodology	38				
3.1	Finit	e element (FE) analysis using ABAQUS	38				
3	3.1.1	ABAQUS/CAE simulations	39				
3	3.1.2	Comparative Selection of FE Analysis Modules	40				
3.2	Func	lamental Continuum Theory	41				
3.2.1		The von Karman shell theory for flat plates and membranes					
3	3.2.2	Dislocation Mechanics	42				
3.3	Mole	Molecular dynamics (MD) simulations					

3.4	Refer	ences	47				
Chapt Nanor	ter 4: membi	Quantitative Analysis and Predictive Engineering of Self-rolling ranes Under Anisotropic Mismatch Strain	of 49				
4.1	Abstr	act	50				
4.2	Intro	duction	51				
4.3. Meth		odology	53				
4.	3.1	Analytical formulation	53				
4.3.2		Roll-up geometries from numerical simulations					
4.4	Resu	ts and Discussion					
4.	4.1	FE simulated and analytically predicted roll-up curvatures	60				
4.	4.2	Implications to practical applications	62				
4.5	Conc	lusion	73				
4.6	Ackn	owledgement	74				
4.7	Supp	orting information	75				
4.7.1		Derivation details for analytical formulation in Section 4.3.1					
4.7.2		Derivation details for the third layer deposition in Section 4.4.2	78				
4.8	Refer	ences	81				
Chapt	ter 5:T	opological patterning effects on the self-rolling of nanomembranes	84				
5.1	Abstr	act	85				
5.2	Intro	luction	86				
5.3	Meth	odology	87				
5.3.1		Analytical formulation	87				
5.	3.2	Numerical simulations	91				
5.4	Resu	lts	92				
5.	4.1	Effect of patterning on roll-up curvature for unidirectional rolling	92				
5.	4.2	Flexible tuning of rolling direction and roll-up geometry via patterning	95				
5.5	Conc	lusion	. 101				
5.6	Acknowledgement		102				
5.7	Supp	orting information	103				
5.8	8 References						
Chapt	ter 6:	Misfit dislocation induced strain relaxation effect on self-rolling of str	ain-				
	A hate	nanomemoranes	110				
0.1	Introduction 1						
0.2	muoo	JUCHOII	. 111				

6.3	Computational Methodology					
6.4	Results and Discussion					
6.4.1 Effects of misfit dislocation on strain gradier		Effects of misfit dislocation on strain gradient of nanomembranes	116			
6.4.2		Effects of misfit dislocation on the rollup curvature of nanomembranes	119			
6.5	Conc	lusion	123			
6.6	Acknowledgement					
6.7	References					
Chap	ter 7:	Effects of material heterogeneity on self-rolling of strain eng	ineered			
memb	oranes		128			
7.1	Absti	ract	129			
7.2	Introduction					
7.3	Methodology					
7.	3.1	Analytical formulation	131			
7.	3.2	Molecular dynamics simulations	136			
7.	3.3	Numerical simulations	136			
7.4	Resu	Its and discussions	137			
7.5	Conclusion					
7.6	Acknowledgement					
7.7	Reference					
Chap	ter 8: (Conclusions	146			
8.1	Majo	r conclusions and implications	146			
8.2	Contribution to the original knowledge					
8.3	Future work					
8.4	References					

List of Figures

Figure 2.2. Scanning electron microscope (SEM) images of the rollup structures (a) an array of rolled-up SiO/SiO₂ nanomembranes [18]. (b) The microtube of $In_xGa_{1-x}As/GaAs$ when sacrificial layer is being etched [45]. (c) The SEM image of strain induced wrinkles [42]. (d) The SEM image of the self rolled up nanospring [46]. [Figures adapted with permissions] ...10

Figure 2.5. Phase diagram of preferential rolling modes (long edge rolling or short edge rolling) for a rectangular strained nanomembranes depending on various wrinkle amplitude and strain gradient. Insets show the snapshots of the rolling up of metallic film made of Cu_{0.59}Ni_{0.40} Mn_{0.01} alloy on the wrinkled film from video microscopy [47]. [Figures adapted with permissions] 13

Figure 2.6. Comparison between experimental measured rollup diameters and the predicted values from Timoshenko formula with assumptions of oxidization layer and surface stress imbalance. (a) Experimental and theoretical results of InAs/GaAs system with additional assumptions of oxidization layer and equal Yong's modulus. The bottom plot indicates the root mean square (RMS) roughness of surface before etching [64]. (b) Experimental and theoretical

Figure 2.9. Atomistic simulations of self-rolled-up structures. (a) Hydrogenation assisted graphene self-rolling into a nanocage [72]. (b) Surface reconstruction effects on self-rolling behaviors of strained bilayer for (001) Ge/Si with (Left) an even number (4 Ge/4 Si) and (Right) an odd number (4Ge/5 Si) of atomic layers [73]. [Figures adapted with permissions]............20

Figure 2.11. The derived resonant mode and sensing capability for microtubes with refractive index $n_2 = 2$, m = 40 and different wall thickness Δ . (a) Wavelengths λ_r (b) *Q*-factors, (c) sensitivities *S* and (d) figures of merit *QS*. The symbols in (a) and (c) are approximated results obtained from waveguide approximation approaches [84]. [Figures adapted with permissions]

Figure 4.1. (a) Schematic illustration of a strained rectangular bilayer nanomembrane of length *L* and width *W*, with mismatch strain ε_0 and $\eta \varepsilon_0$ along the x_1 (longitude) and x_2 (transverse) directions respectively. The top and bottom layers are respectively of thicknesses (h_t , h_b),

Figure 4.4. Comparison between theoretically predicted (lines) and experimentally measured (open symbols) roll-up diameters for the (a) $GaAs^{(top)}/InAs^{(bottom)}$ nanomembrane [32] and (b) $SiN_x^{(top)}/SiN_x^{(bottom)}$ nanomembrane [21], as the thickness of the top layer varies. The parameter η denotes the anisotropy of lattice mismatch. The predictions from Timoshenko formula (dotted lines) and the previous study by Grunmann [29] (pink dash-dot-dot line in (a)) are also included for comparison.

Figure 4.7. Schematic illustration of the role of corner attachment in achieving controlled unidirectional roll-up along the longitude direction, for nanomembranes. The figures in left and right columns present the initial and resultant FE-simulated roll-up configurations respectively, showing the involvement of transverse rolling mode for (a) rectangular and (c) U-shape nanomembranes without corner attachments, and the realization of desirable longitude roll-up for (b) rectangular and (d) U-shape nanomembranes (See videos in Electronic Supplementary Information). The red segments in (c) and (d) indicate the clamped regions in U-shape nanomembranes.

Figure 5.2. Comparison between the FE simulated (symbols) and predicted (lines, from Eqs. 5.6-9 rolling curvature κ_{11} as the pattern thickness H_p and pattern densities (ρ_1 , ρ_2) vary, for

Figure 5.3. Comparison between the FE simulated (symbols) and predicted (lines, from Eqs. (5.6-9)) rolling curvature κ_{11} as the pattern thickness H_p and pattern densities (ρ_1 , ρ_2) vary, for the GaN^(top)/AlN^(bottom) nanomembrane system with (a) grating and (b) rectangular patterns.

Figure 5.6. Schematic illustration of (a) strips cut from a grating-patterned host nanomembrane, aligned at different angles α (e.g., $\alpha = 37^{\circ}$, 45° and 57°) to the direction perpendicular to the grating pattern, which, after roll-up (with the rolling direction indicated by the black dashed arrows in (a)), results in (b) helixes with axis parallel to the grating pattern (obtained from FE simulations). (b) FE simulated helical strips with designed helical angles ($\alpha = 37^{\circ}, 45^{\circ}, 57^{\circ}$). (c) and (d) plot the designed helix diameter D_{pred} and pitch period P_{pred} (from analytical predictions) against the ones (i.e., denoted as D_{sim} and P_{sim} respectively) obtained from FE simulations, showing excellent agreement. In this illustrative example, the nanomembrane

Figure 6.1. Three dimensional schematic illustrations of the (a) zinc blend CdS/CdTe_{0.5}S_{0.5} and (c) wurtzite GaN/In_{0.5}Ga_{0.5}N where the shaded plane indicated the interface. (b) and (d) Projection views of atomic configurations along [011] and [$11\overline{2}0$] to illustrate the local atom arrangements on the interface. Cd, Te and S atoms are colored in blue, red and yellow, respectively. The N atoms are indicated by small blue spheres, while In and Ga atoms are represented by purple and golden spheres.

Figure 6.8. The predicted and MD simulated rolling diameter as a function of the misfit dislocation density for (a) CdTe/CdTe_{0.5}S_{0.5} and (b) GaN/In_{0.5}Ga_{0.5}N, respectively......123

 Figure 7.1.
 Schematic illustration of rectangular strained bilayer membranes with aligned

 heterogeneous elements colored in the blue.
 132

Figure 7.3. The simulated geometry of the $CdTe_xS_{1-x}/CdTe$ system where Cd, Te and S atoms are colored red, blue and light yellow respectively. (b) The MD simulated rolling diameter as a function of the thickness for the self-rolling of CdS/CdTe and CdTe_0.5S_0.5/CdTe systems. ...138

Figure 7.4. The representative self-rolling models with alternative distributed strip structures of CdTe_{0.4}S_{0.6}/CdTe (a) and (b) indicate the longitudinal and lateral alternative distributed strip structures, respectively, while (c) and (d) represent the corresponding equilibrium geometries.

List of Tables

Table	5.1.	The	relevant	material	properties	of	the	two	nanomembrane	systems,
GaAs ^{(to}	^{op)} /Ino.:	2Ga0.84	As ^(bottom) and	nd GaN(top))/AlN ^(bottom)	[22, 3	53, 56	6] cons	sidered in the pres	ent study.
										93
Table	6.1. T	he red	uced elast	ic stiffness	ses obtained	for	CdTe	/CdTe	co.5So.5 and GaN/In	10.5Gao.5N 122
Table '	7.1. TI	ne valı	ues of elas	tic constan	nts used in th	e M	D sin	nulatic	ons	

Chapter 1: Introduction

Pioneered by Schmidt and Melechko [1, 2], the self-rolling technology of strain engineered nanomembranes, which combines top-down and bottom-up approaches, has been stimulating increasing interest for various electrical and optical devices [3-5]. The self-rolling technology involves the deposition of differentially strained nanomembranes of bi and multistacked structures on a removable sacrificial layer for subsequent release and relaxation, which leads to deterministically rearranging the geometry of nanomembranes and fabrication of folded or tubular spiral configurations including tubes [6], helices [7], rings [8], wrinkles [9] and other advanced microarchitectures, having diameters ranging from a few nanometers to several hundred microns, and wall thicknesses on the magnitude of nanometers. This way of creating 3D structures capable of programmable shape transformations [10, 11] and to be fabricated from, in principle, any kind of functional materials, promising unprecedented possibilities for nanofabrication approaches [12, 13].

These novel and unique programmable rolled-up structures, combined with a plentiful pool of available functional materials, have driven the emerge of numbers of research fields in on-chip lasing [14], optical communication [15], energy storage [16], metamaterials [17, 18] etc., by employing the rolled-up geometries as key components, providing a facile path towards the design of tunable nanodevices. For instance, by large area rolling-up ultracompact nanomembranes being metallic or dielectric, capacitors arrays and field effect transistors have been demonstrated with tailorable electrical properties [19]. The development of self-rolling technology has also proved its feasibility of fabricating the steerable tubular microrockets/microrobots that thrust out bubbles to drive the micro-objects moving while employing external electric/magnetic fields as steering [20, 21]. Additionally, the rolled-up

microtube could be adopted as an excellent platform for the integrations of bio- and microfluidic components into microelectronic sensors because of tubular structures naturally acted as a fluidic handling channel [22, 23]. Furthermore, the rolled-up tubular optical cavities have been applied as whispering gallery mode (WGM) of the optical ring resonators combining with diversity materials of tunable refractive index to achieve high quality factor, low mode volume, and large optical density [24-28]. In those applications spanning various fields, the precise geometrical engineering of 3D structures resulting from self-rolling technology has been considered as the cornerstone for manufacturing nanodevices with tunable properties, which requires the deeply understanding of the self-rolled-up mechanics of strain engineered nanomembranes.

The objective of this thesis is to provide mechanistic insights and develop theoretical frameworks toward accurate geometry manipulation of rollup nanomembranes. To achieve the above goal, the mechanics and dynamics during the self-rolled-up process of strain engineered nanomembranes were systematically investigated accounting for the influences of anisotropic strain, surface patterning, material inhomogeneity and misfit dislocations on the resultant sophisticated rollup configurations. The thesis specifically focuses on the following aspects:

a) Analytical modeling the bi-directional rollup of strained nanomembranes to investigate the influences of anisotropic lattice mismatch on the rollup curvature and directions of nanomembranes;

b) Examining the effects of surface patterning on the curvature of rolled-up nanomembranes and patterning induced controllable self-helixing processes;

c) Quantifying the strain relaxation effects induced by misfit dislocations on the selfrolling of nanomembrane using atomistic simulations combined with continuum theory;

d) Investigating the self-rolling process of strained nanomembranes with heterogeneous elements embedded.

The major conclusions obtained from the thesis work are presented in Chapters 4-7. Specifically:

• Chapter 4 quantitatively assessed the effect of anisotropic mismatch strain on the rolledup curvature of nanomembranes employing continuum theory and finite-element analysis. Two strategies, namely third-layer deposition and corner geometry engineering, were proposed to predictively manipulate the bidirectional rolling competition of strained nanomembranes to achieve improved roll-up yield. In particular for the strategy of corner engineering, microfabrication experiments have been performed to showcase its practical application and effectiveness.

• Chapter 5 proposed a quantitative modeling framework for investigating of the effects of topographic patterning on the rolling direction and curvature of rolled-up nanomembranes. The accuracy and reliability of our theoretical model has been demonstrated by its application to helix-shape structures to showcase a quantitative route to achieve controllable helical rolling of nanomembranes through strip patterning, showing excellent agreement with both experimental observations and finite-element simulations.

• Chapter 6 presented a combined model that bridged atomistic information of dislocation distortion with continuum theory, and quantitatively evaluated the dislocations effects on the self-rolling of strain-engineered nanomembranes. The accuracy and effectiveness of theoretical model have been verified via two representative material systems including (GaN/In_{0.5}Ga_{0.5}N) of wurtzite lattice, and II–VI materials (CdTe/CdTe_{0.5}S_{0.5}) of zinc-blend lattice.

• Chapter 7 clarified the dependence of self-rolling behaviors of strained membranes on the distribution and percentage of heterogeneously embedded elements, validated at macro and microscopic scales through molecular dynamics (MD) simulations and finite element analysis.

1.1 References

[1] O.G. Schmidt, K. Eberl, Nanotechnology: Thin solid films roll up into nanotubes, Nature, 410 (2001) 168-168.

[2] A.V. Melechko, V.I. Merkulov, T.E. McKnight, M.A. Guillorn, K.L. Klein, D.H. Lowndes, M.L. Simpson, Vertically aligned carbon nanofibers and related structures: Controlled synthesis and directed assembly, J Appl Phys, 97 (2005).

[3] J.X. Li, W.J. Liu, J.Y. Wang, I. Rozen, S. He, C.R. Chen, H.G. Kim, H.J. Lee, H.B.R. Lee, S.H. Kwon, T.L. Li, L.Q. Li, J. Wang, Y.F. Mei, Nanoconfined Atomic Layer Deposition of TiO2/Pt Nanotubes: Toward Ultrasmall Highly Efficient Catalytic Nanorockets, Advanced Functional Materials, 27 (2017).

[4] Q.L. Guo, G. Wang, D. Chen, G.J. Li, G.S. Huang, M. Zhang, X. Wang, Y.F. Mei, Z.F. Di, Exceptional transport property in a rolled-up germanium tube, Appl Phys Lett, 110 (2017).

[5] W.P. Si, I. Monch, C.L. Yan, J.W. Deng, S.L. Li, G.G. Lin, L.Y. Han, Y.F. Mei, O.G. Schmidt, A Single Rolled-Up Si Tube Battery for the Study of Electrochemical Kinetics, Electrical Conductivity, and Structural Integrity, Adv Mater, 26 (2014) 7973-7978.

[6] I.S. Chun, V.B. Verma, V.C. Elarde, S.W. Kim, J.M. Zuo, J.J. Coleman, X. Li, InGaAs/GaAs 3D architecture formation by strain-induced self-rolling with lithographically defined rectangular stripe arrays, J Cryst Growth, 310 (2008) 2353-2358.

[7] G.S. Huang, Y.F. Mei, Helices in micro-world: Materials, properties, and applications, J Materiomics, 1 (2015) 296-306.

[8] M.H. Huang, F. Cavallo, F. Liu, M.G. Lagally, Nanomechanical architecture of semiconductor nanomembranes, Nanoscale, 3 (2011) 96-120.

[9] T. Ma, H.S. Liang, G. Chen, B. Poon, H.Q. Jiang, H.B. Yu, Micro-strain sensing using wrinkled stiff thin films on soft substrates as tunable optical grating, Optics Express, 21 (2013) 11994-12001.

[10] T. van Manen, S. Janbaz, A.A. Zadpoor, Programming 2D/3D shape-shifting with hobbyist 3D printers, Materials Horizons, (2017).

[11] N. An, M.E. Li, J.X. Zhou, Predicting origami-inspired programmable self-folding of hydrogel trilayers, Smart Mater Struct, 25 (2016).

[12] Y.F. Mei, G.S. Huang, A.A. Solovev, E.B. Urena, I. Moench, F. Ding, T. Reindl, R.K.Y. Fu, P.K. Chu, O.G. Schmidt, Versatile Approach for Integrative and Functionalized Tubes by Strain Engineering of Nanomembranes on Polymers, Adv Mater, 20 (2008) 4085-4090.

[13] Y.H. Zhang, F. Zhang, Z. Yan, Q. Ma, X.L. Li, Y.G. Huang, J.A. Rogers, Printing, folding and assembly methods for forming 3D mesostructures in advanced materials, Nat Rev Mater, 2 (2017).

[14] A. Madani, M. Kleinert, D. Stolarek, L. Zimmermann, L.B. Ma, O.G. Schmidt, Vertical optical ring resonators fully integrated with nanophotonic waveguides on silicon-oninsulator substrates, Optics Letters, 40 (2015) 3826-3829.

[15] S. Bottner, M.R. Jorgensen, O.G. Schmidt, Rolled-up nanotechnology: 3D photonic materials by design, Scripta Mater, 122 (2016) 119-124.

[16] X.F. Wang, Y. Chen, O.G. Schmidt, C.L. Yan, Engineered nanomembranes for smart energy storage devices, Chem Soc Rev, 45 (2016) 1308-1330.

[17] L.Z. Xu, T.C. Shyu, N.A. Kotov, Origami and Kirigami Nanocomposites, Acs Nano, 11 (2017) 7587-7599.

[18] A.S. Gladman, E.A. Matsumoto, R.G. Nuzzo, L. Mahadevan, J.A. Lewis, Biomimetic 4D printing, Nat Mater, 15 (2016) 413-418.

[19] R. Sharma, C.C.B. Bufon, D. Grimm, R. Sommer, A. Wollatz, J. Schadewald, D.J. Thurmer, P.F. Siles, M. Bauer, O.G. Schmidt, Large-Area Rolled-Up Nanomembrane Capacitor Arrays for Electrostatic Energy Storage, Adv Energy Mater, 4 (2014).

[20] H. Wang, M. Pumera, Fabrication of Micro/Nanoscale Motors, Chem Rev, 115 (2015) 8704-8735.

[21] Y.F. Mei, A.A. Solovev, S. Sanchez, O.G. Schmidt, Rolled-up nanotech on polymers: from basic perception to self-propelled catalytic microengines, Chem Soc Rev, 40 (2011) 2109-2119.
[22] D. Grimm, C.C. Bof Bufon, C. Deneke, P. Atkinson, D.J. Thurmer, F. Schäffel, S. Gorantla,

A. Bachmatiuk, O.G. Schmidt, Rolled-up nanomembranes as compact 3D architectures for field effect transistors and fluidic sensing applications, Nano letters, 13 (2012) 213-218.

[23] C. Deneke, O. Schmidt, Real-time formation, accurate positioning, and fluid filling of single rolled-up nanotubes, Applied Physics Letters, 85 (2004) 2914-2916.

[24] J. Wang, T.R. Zhan, G.S. Huang, P.K. Chu, Y.F. Mei, Optical microcavities with tubular geometry: properties and applications, Laser Photonics Rev, 8 (2014) 521-547.

[25] S.L. Li, L.B. Ma, H.L. Zhen, M.R. Jorgensen, S. Kiravittaya, O.G. Schmidt, Dynamic axial mode tuning in a rolled-up optical microcavity, Appl Phys Lett, 101 (2012).

[26] S.W. Tang, Y.F. Fang, Z.W. Liu, L. Zhou, Y.F. Mei, Tubular optical microcavities of indefinite medium for sensitive liquid refractometers, Lab Chip, 16 (2016) 182-187.

[27] X.L. Li, Self-rolled-up microtube ring resonators: a review of geometrical and resonant properties, Adv Opt Photonics, 3 (2011) 366-387.

[28] G.S. Huang, Y.F. Mei, Electromagnetic wave propagation in a rolled-up tubular microcavity, J Mater Chem C, 5 (2017) 2758-2770.

Chapter 2: Literature Review

2.1 Rolled-up nanotechnology

For decades, advances in fabrication methods of controllable 3D micro-and nanostructures and their ordered assemblies have garnered significant interest, driven by their great technological relevance and potential for creating material systems of novel properties and unprecedented functionalities either individually or collectively [1-6]. These fabrication methods can be generally divided into two categories: the top-down (template-free) and bottomup (template-assisted) approaches [7-9]. The top-down approaches are predominantly lithography-based procedures, including photolithography [10], scanning lithography [11], soft lithography [12], and nanocontact printing [13], among others. The bottom-up route involves templated deposition growth such as chemical vapour deposition [14] and molecular beam epitaxy [15], combined with the subsequent self-assembly of components into more complex architectures [16]. Self-rolling strain-engineered nanomembranes on basis of the bottom-up method is gaining increasing interest due to the unique mechanical flexibility and geometrical variabilities along with excellent machinability from micro to nanoscale [17-20]. It is perfectly compatible with the top-down patterning technology through photolithography, offering great technological capability. For example, it can enable the production of numerous identical threedimensional (3D) geometries without suffering from the stochastic variation in nanostructure parameters like size, shape, position, and composition [21-24]. Meanwhile, it is also very convenient to accurately integrate the self rolled-up structures on a chip [25]. Therefore, the strain induced self-rolling of nanomembranes is expected to play increasingly important roles in various potential applications in the microelectronics industry [26-28]. For instance, in terms of smart energy storage devices, the 3D tubular nanomembranes fabricated through the strain induced self-rolling have been designed and explored extensively for the on-chip integration of fast-micro-lithium storage, due to its chemical stability, large active surface area, and high energy and power densities [29-31]. The self-rolling technique offers an inexpensive and accessible route to fabricate helical ribbon micromotors, actuated by either an electric field or a rotating magnetic field, which allows the micromotors to capture and release preselected carriers for biomedical applications [32, 33]. Also, the rollup nanomembranes have been extensively studied for creating the whispering-gallery mode (WGM) microcavity for sensing applications with features of high quality factor, low mode volume, and large optical density [34-36]. However, despite self-rolling strain-engineered nanomembranes being widely applied in optoelectronic devices and (MEMS/NEMS) systems, its potential is significantly limited due to inadequate understanding of the mechanisms underlying the self-rolling process. Critical knowledge regarding the mechanics and dynamics during the self-rolled-up process of strain engineered nanomembranes remain missing, which makes it difficult to control geometries of the resultant nanostructures and integrate state-of-the-art functionality required by those applications, and thus necessitate further investigation.

2.2 Generic design method of self-rolled-up structures

2.2.1 Mechanisms of strain induced self-rolling of nanomembranes

Precise control and design of self-rolled-up nanomembranes rely on the deep understanding of their fabrication principles and related mechanical mechanisms. The generic fabrication approach for creating strain induced self-rolling structures consists of several essential fabrication steps, summarized as following:

As shown in Figure 2.1, the materials with predefined geometry are first prepared based on a 2D template through the nanofilm deposition/growth [37, 38]. The material system includes (1) a sacrificial layer that can be selectively etched off from the substrate, and (2) a strained bilayer or multilayer on the top of the sacrificial layer consisting of materials of different lattice constants or a single layer with certain internal strain gradient. The sacrificial layer is then selectively removed, and subsequently the strained bilayer or multilayer would gradually detach from the substrate. As the strained layer detaches, the release of the stored strain energy drives the layer into rolling, bending, twisting, buckling or other forms of geometrical transformation. This leads to the designed 2D patterns taking different final 3D configurations, like tubes [39, 40], helix [41], and periodic wrinkles [42] depending on the elastic properties and thicknesses of the strained layers as well as the initial geometric conditions defined by the lithographic process. Fig. 2.2 presents a few experimental examples of nanostructures resulted from self-rolling of strained nanomembranes. It's worth noting that based on the same mechanical principle, the built-in strain may also respond to external factors, including pH, heat, light density, or temperature for certain material systems (e.g., stimuli-responsive hydrogels) [43]. Therefore, the geometrical transformation of nanomembranes could be reversibly controlled to produce, e.g., a shape-memory effect that has significant potential for creating a wide range of stimuli-responsive structures for the smart deployable devices [44].



Figure 2.1. Schematic illustrating the process of strain-driven rolling up. (a) The formation, via deposition, of a strained bilayer nanomembrane on a substrate with a sacrificial layer. (b) Rolling of the strained bilayer nanomembrane as the sacrificial layer is etched, driven by the releasing of built-in strain energy.



Figure 2.2. Scanning electron microscope (SEM) images of the rollup structures (a) an array of rolled-up SiO/SiO₂ nanomembranes [18]. (b) The microtube of $In_xGa_{1-x}As/GaAs$ when sacrificial layer is being etched [45]. (c) The SEM image of strain induced wrinkles [42]. (d) The SEM image of the self rolled up nanospring [46]. [Figures adapted with permissions]

2.2.2 Rolling direction competition for rectangular nanomembranes

It should be noted that there exists competition among all potential self-rolling directions as the strained nanomembranes are detached form the substrate. Such competition, demonstrated in Fig. 2.3, naturally derives from the simultaneous release of strain energy along different directions. It creates a critical challenge for achieving precise control of the rolling configurations.



Figure 2.3. SEM snapshots of (a) 30s and (b) 45s illustrating the competition between roll-up directions of the rectangle strained $In_xGa_{1-x}As/GaAs$ structures [39]. [Figures adapted with permissions]

Extensive research effort [39, 47-51] have been devoted to the control of rolling directions. Earlier studies reveal that the dominant self-rolling direction and final geometrical morphology after self-rolling greatly depend on the initial geometric conditions, the history of the rolling process, presence of surface patterns and elastic moduli of strained nanomembranes. For instance, it has been demonstrated that rectangular strained nanomembranes constituted by elastically isotropic materials with large width/length ratios tend to start (and continue) rolling from the long edge side and form a tubular geometry [39]. This preferential bending along the long edge side was attributed to the existence of doubly curved regions at the curled edges that lowers the energy of the whole system and the energy difference between two rolling modes, namely, long edge rolling and short edge rolling, increases with the width and length ratio of strained bilayers [52]. Besides, the dominant rolling direction could be determined by the combination of edge effects and the most compliant direction when the strained nanomembranes being elastically anisotropic, and it thus allows a long narrow strip to be transformed into a helical shape depending on the angle between the alignment direction of the strip and its most compliant direction, as illustrated in Fig 2.4 (a) [46, 53]. The three geometric

parameters, such as the helical angle θ , the helical radius *R*, and the pitch period *p*, satisfy the condition of $\tan \theta = \frac{P}{\pi D}$. Additionally, patterned grating structures on the surface of strained metal bilayer system have been proposed to deterministically control the self-rolling direction to facilitate the fabrication of helical structured micro-springs [50]. And the rolling direction of the bilayer system is indicated to be always perpendicular to the grating structures and could be controlled by adjusting the pattern angle of the grating structures on the strained nanomembranes, as demonstrated in Fig. 2.4 (b). In addition, a fabrication strategy based on manipulating the gradient strain and initially wrinkled surface have also developed to achieve the precise control of the preferential rolling direction for the strained nanomembranes in rectangular shape. The phase diagram of the preferential rolling direction is given in Fig. 2.5, depending on the strain gradient and wrinkle amplitudes.



Figure 2.4. Two methods that control the dominant rolling up direction of strained nanomembranes to create the helical geometry. (a) Employing the elastic compliant direction and edge effects to construct the helix with diameter *d*, pitch *p*, and helicity angle θ , where [*xyz*] represents the crystal orientation of the stripe, while [*uvw*] denotes the scrolling direction [46]. (b) A process flow of using surface imprint patterns to define rolling direction of strained Ti film to design the grating-structured metallic micro springs [50]. [Figures adapted with permissions]



Figure 2.5. Phase diagram of preferential rolling modes (long edge rolling or short edge rolling) for a rectangular strained nanomembranes depending on various wrinkle amplitude and strain gradient. Insets show the snapshots of the rolling up of metallic film made of Cu_{0.59}Ni_{0.40} Mn_{0.01} alloy on the wrinkled film from video microscopy [47]. [Figures adapted with permissions]

2.2.3 Controllable curvature of self-rolled-up structures

Besides the control of rolling directions of strained nanomembranes, another critical component to achieve precise design of rolled-up configurations of strained nanomembranes is curvature manipulation. The curvature variation of rolled-up nanostructures allows controllable modification of physical properties of systems, and nowadays the correlation between curvature geometry of rolled-up structures and physical properties of devices has become the main topic of extensive research in various areas of physics, particularly for the low-dimensional systems [54-56]. Balhorn et al. showed the modifications of mode spectrum of ferromagnetic Microtubes could be achieved through tailoring the tube radius and number of rolled-up layers [57]. Streubel et al. revealed that curvature adjustment can be used as a means to tune magnetic properties, including magnetic equilibrium states, magnetostatic interaction and magnetic domain patterning, which can enable the cylindrically curved magnetic architectures to promise applications for data storage and magnetic access memory devices [58]. Furthermore, it was

also demonstrated that electronic transport properties can be engineered by varying the curvature [59].

Generally, the curvature of the final 3D geometry resulted from self-rolling of a strained nanomembrane is determined by the mismatch strain and/or strain gradient of the nanomembrane. For a simple strained bilayer system, the variation of the curvature can be roughly evaluated using the following equation based on the macroscopic continuum mechanics model developed by Timoshenko where a slender elastic plate has been considered comprising two strained layers [60]. The deformation of three-dimensional plate is approximately represented using the deformation of the mid-plane of plate in two dimensions employing the Kirchhoff shell assumptions [61, 62]. Eq. (2.1) gives the expression of bending curvature of rolled-up plate, where *E* and *a* represent the elasticity modulus and the layer thickness, respectively. The subscripts *t* and *b* are used for the label indices of top and bottom layers. *h* denotes the total thickness ($h = h_t + h_b$), ε is the mismatch strain of the bilayer system, and κ is the rolling curvature of deformed nanomembrane.

$$\frac{1}{\kappa} = \frac{h\left(3(1+m)^2 + (1+mn)\left(m^2 + \frac{1}{mn}\right)\right)}{3(\varepsilon_t - \varepsilon_b)(1+m)^2}, m = \frac{h_t}{h_b}, n = \frac{E_t(1-\nu_b)}{E_b(1-\nu_t)}$$
(2.1)

The detailed derivation of this formula is on the basis of energy minimization principle, in which two uniform curvatures ($\kappa_{11} = \kappa_{22} = \kappa$) are assumed for the rolled-up nanomembranes for simplicity of analytical treatment. The strain components in the mid-plane (at height h/2 above the base of the bottom layer) are considered to be equal and isotropic ($\gamma_{11} = \gamma_{22} = \gamma$). Therefore, the strain energy density in the top and bottom layer can be determined as

$$U_t = \frac{1}{2}\sigma_{ij}^t \varepsilon_{ij}^t = \frac{E_t}{2 \cdot (1 - \nu_t)} (\gamma + \kappa \cdot x_3)^2$$
(2.2)

$$U_b = \frac{1}{2}\sigma_{ij}^b \varepsilon_{ij}^b = \frac{E_b}{2 \cdot (1 - \nu_b)} (\gamma + \kappa \cdot x_3 + \varepsilon)^2$$
(2.3)

Where ε_{ij}^t and ε_{ij}^b are the strain components in the top and bottom layer, respectively. σ_{ij}^t and σ_{ij}^b are corresponding stress components. x_3 represents the distance from the mid-plane. Therefore, the total potential energy *U* can be further calculated by integration Eqs (2-3) over the whole system,

$$U = \iiint_V \left(\frac{1}{2}\sigma_{ij}^b \varepsilon_{ij}^b + \frac{1}{2}\sigma_{ij}^t \varepsilon_{ij}^t\right) dV$$
(2.4)

Finally, the equilibrium values of γ and κ can be obtained by solving the linear equations derived from $\frac{\partial U}{\partial \gamma} = 0$ and $\frac{\partial U}{\partial \kappa} = 0$. Here, it should be emphasized that the presumptions exclude the possibility of anisotropic rolling curvature and initial mismatch strain anisotropy, and thus fail to elucidate the multi-stable equilibrium of self-rolling behaviors.

In addition, it mainly focuses on initially flat strained nanomembranes on regular substrates, and neglects the influence of initial curved substrate. The investigation of self-rolling architectures with adjustable bidirectional curvature remains a huge challenging. Actually, considerable deviations in rollup curvature exist between model predictions and experimental measurements. For example, the study by Zhang et al. [63] gives a tube diameter of 2.15µm for the rolled up Si/SiGe bilayer, whereas 2.44 µm is obtained from Eq. (2.1). In the study by Chun et al. on GaAs/In_{0.2}Ga_{0.8}As system [45], the diameter of the self-rolled-up structure was measured to be 884 nm, while 1024 nm is predicted by the Timoshenko formula (~14% overestimation). Such discrepancy between experimental measurements and theoretical predictions was also confirmed by other studies [64, 65] The discrepancy has been attributed to different factors, such as nonlinear strain effect [66], assumption of additional mismatch strain or oxidation layers [64, 67], surface tension and reconstruction [65, 68]. For example, the effects of oxidation layers and surface tension are illustrated in Fig. 2.6. However, those efforts in explaining the discrepancy involved additional assumptions, and are mostly qualitative with notable inconsistency [49, 69, 70]. For instance, the self-rolling induced by the intrinsic surface-
stress imbalance can only be applied into systems of several atomic layers in thickness, where surface dimers are relocated to have gradient stress along the thickness direction. On the other hand, this formula is not suitable for thick layers of strained nanomembranes or highly mismatched systems like AlN/GaN. This is due to the inaccurate evaluation of mismatch strain, where the built-in strain gradient would be reduced through the nucleation of interface misfit dislocations and the term of mismatch strain in Eq (2.1) should be corrected to include the effects of dislocations. The research regarding the self-rolling of ultrathin AlN/GaN porous crystalline nanomembranes has confirmed the existence of dislocations in the rolled up microtubes, as shown in Fig.2.7 [71]. The basic mechanism of dislocation nucleation in strained curved nanomembranes can be explained in the framework of continuum mechanics during the deposition process of heterogeneous nanomembranes. The elastic strain energy resulting from the lattice mismatch is released in the form of surface energy during the initial deposition stage, producing dislocation-free, equally spaced islands when the island size is small [42]. However, as the island continues to expand, the increasing surface energy cannot be compensated by the decrease in the strain energy caused by the lattice mismatch. As the island volume increase, the strain energy raises, eventually resulting in the nucleation of dislocations in the boundary of islands. With the nucleation of dislocations, the islands are no longer coherently strained, leading to heterogeneity in the bilayer system. The nucleated dislocations would relax the mismatch strain to some degree, and the further quantitively evaluation at atomistic scale for this strain relaxation need to be performed to examine the corresponding curvature variation.



Figure 2.6. Comparison between experimental measured rollup diameters and the predicted values from Timoshenko formula with assumptions of oxidization layer and surface stress imbalance. (a) Experimental and theoretical results of InAs/GaAs system with additional assumptions of oxidization layer and equal Yong's modulus. The bottom plot indicates the root mean square (RMS) roughness of surface before etching [64]. (b) Experimental and theoretical results of Si/SiGe system with surface stress modifications [65]. [Figures adapted with permissions]



Figure 2.7. The self-rolled-up microtube with dislocations embedded (a) and (b) SEM images of microtubes made from strained AlN nanomembrane (20 nm thick) on Si substrate. The insets show the enlarged SEM images of the open end of microtube (c) The enlarged TEM of the rollup wall showing the pores with a size of 10~20 nm; (d) The fast Fourier transformed filtered image of strained AlN layer to test crystal defects and the lattice spacing (e) The magnified TEM image for the rectangle region in (d) and the lattice spacing several dislocations are labelled by the black arrows [71]. [Figures adapted with permissions]

2.2.4 Computational modeling of self-rolled-up structures

For complex multilayered structures with predefined surface functional patterns for electronics and optics applications, numerical method is preferred to predict the rolling curvature and understand the kinetics involved in the rolling process, because the nonuniform strain gradient along the thickness direction and the surface strain imbalance induced by the distribution of patterns are difficult to be incorporated into the analytical method. Therefore, simulations based on finite element method (FEM) have been proposed to study the rolling process of strain engineered nanomembranes, focusing on the size effects on the final rolling competition and the resultant structures [39], and precision curvature design of different coaxial inner diameters [48]. In the FEM simulations, the transient quasi-static method with moving boundary conditions is usually employed to explore the possible 3D rollup configurations. During the releasing process, the dynamic etching of strained nanomembranes is modelled through slow boundary movement, while the internal mismatch strain is achieved by assigning different thermal expansion coefficients to each layer and temperature variation. Fig. 2.8 shows the structural design of self-rolled-up nanomembranes using silicon nitride based on the FEM simulations. An excellent agreement between experimental results and numerical simulations has been observed for the fabrications of coaxial tubular geometry and double helix structures.



Figure 2.8. Structural design of self-rolled-up nanomembranes based on the FEM simulations. (a) Experimental fabricated coaxial tubular geometry with two diameters controlled by the local thickness adjustment. The diameters for the first and second turn are meassured to be 6.66 μ m and 13.71 μ m, respectively. (b) FEM simulated tubular structure with diameters 7 and 14 μ m. (c) Experimental and simulated double helix structures resulting from a patterned origami planar configuration, achieved by the anisotropic release process [48]. [Figures adapted with permissions]

It is important to note that the driving force of the self-rolling behaviors may also originate from selective molecular adsorption [72], and the imbalance of surface stress when the thickness of strained bilayer is reduced to a few atomic layers [73], where continuum approaches fails and experiments alone often cannot obtain key information about the exact atomic arrangement of rolled up structures. Therefore, atomistic simulations are demanded to examine the self-rolling mechanics and related electronic properties. A direct example in Fig. 2.9 (a) shows the transformation processes of planar graphene to 3D nanocage with the assistance of H molecular adsorption, where precise manipulation of the adsorption position of H plays a critical role to fold and stabilize the resultant nanocage for molecular storage. The folding angles of single-sided hydrogenation along the selected edges are determined through

detailed molecular dynamics (MD) simulations. It has been further revealed that this hydrogenation-assisted graphene nanocage could reversibly opened and closed when a programmable external electric field is applied, which is extendable for the application of volumetric light confinement [74]. Fig. 2.9 (b) illustrates the surface reconstruction effects on the self-rolling behaviors of strained Ge/Si bilayer with various atom layers. The MD simulations indicate that the surface stress imbalance due to the surface reconstruction is anisotropic and has additional effects on the principal axis of self-rolling, especially for the case of large surface-to-volume ratio.



Figure 2.9. Atomistic simulations of self-rolled-up structures. (a) Hydrogenation assisted graphene self-rolling into a nanocage [72]. (b) Surface reconstruction effects on self-rolling behaviors of strained bilayer for (001) Ge/Si with (Left) an even number (4 Ge/4 Si) and (Right) an odd number (4Ge/5 Si) of atomic layers [73]. [Figures adapted with permissions]

2.3 Practical applications of strain induced self-rolled-up structures

2.3.1 Self-rolled-up microtube used for microcavity resonators

The self-rolled-up microtubes have shown great potential as components in tubular microcavity resonators with higher quality factors and finer spectral resolution [75, 76]. In the tubular microcavity, light can be defined in the circular structure and distributed by wavelength and distinguished at specific resonant frequencies [77]. The wall thicknesses of such tubular microcavities are smaller than the light resonance wavelength and thus the related resonances are highly sensitive to ambient refractive index [78] and can have strong on-resonance coupling of attached dye emitters to optical modes for the optofluidic sensing and lasing applications [79]. Fig. 2.10 shows the interaction between light and Rhodamine 6G (R6G) molecule. One can note that enhanced photoluminescence and Raman scattering effects have been achieved. It implies that the self-rolled-up microtube can be used as an optical platform for the applications like single nanoparticle detection and optofluidic sensing.



Figure 2.10. The Rolled-up tubular microcavity for the optofluidic sensing and lasing applications. (a) Schematic illustration of a rolled-up tubular microcavity with R6G molecules incorporated. (b) The average electric filed intensify of the microtube has been calculated as a function of wavelengths. The inset on the upper right indicates the related on-resonance field profiles [79]. [Figures adapted with permissions]

The unique advantage of strain induced self-rolled-up microcavity resonators lies in its high freedom in the selection of constituting material, which enables multiple functional materials, like (2D materials or high fluorescence materials) to be incorporated into self-rolled-up structures to achieve enhanced performance [80-82]. In addition, its excellent geometrical flexibility enables the production of microcavities with controllable diameters, wall thickness and refractive index, thus allowing tuning the resonant wavelengths, Q-factor and even optical sensitivity [83-85]. Therefore, to accurately evaluate those properties and connect them with the geometrical configurations of self rolled-up structures, several approaches with rigorous analytical formulas have been proposed, including waveguide approximation approach [86], Mie scattering method [84, 87] and the finite-difference time-domain (FDTD) method [88, 89]. However, the waveguide approximation approaches are oversimplified and not suitable for the case of liquid-in-tube due to the considerable deviation from the experimental results

The numerical simulations using FDTD are quite time consuming. Therefore, the Mie scattering method is generally considered as a reliable and robust way to theoretically investigate the greatest possible performance of self-rolled-up microcavity resonators and develop better tunable optofluidic system. Brief descriptions of Mie scattering method are given below [84, 85, 90]. Considering a microtube made of N strained layers with different refractive indices, when TM (TE) waves are incident on a microtube, the electric field E_z or magnetic field H_z in the *i*-th and (*i*+1)-th layer could be approximated as

$$\Phi^{i} = \sum_{m} \left[a_{i,m} J_{m}(k_{i}r) + b_{i,m} H_{m}^{1}(k_{i}r) \right] e^{im\Phi}$$
(2.5)

$$\Phi^{i+1} = \sum_{m} \left[a_{i+1,m} J_m(k_{i+1}r) + b_{i+1,m} H_m^1(k_{i+1}r) \right] e^{im\Phi}$$
(2.6)

where $k_i = n_i k_0$, n_i represents the refractive index of the *i*-th layer and $k_0 = \frac{2\pi}{\lambda}$. λ indicates the wavelength in the vacuum. The integer m is the order of resonance. Here, cylindrical coordinates (r, Φ) are used and centered on the microtube. The Bessel function J_m and Hankel function of the first kind H_m^1 denote the incident and scattering waves, respectively. Then, the scattering coefficients $D_{i,m} = b_{i,m}/a_{i,m}$ are obtained employing the continuity of E_z and H_z at the interface r=R ($\Phi^i(\mathbf{R}) = \Phi^{i+1}(\mathbf{R})$ and $\frac{\partial \Phi^i(\mathbf{R})}{\partial \mathbf{r}} = \frac{\partial \Phi^i(\mathbf{R})}{\partial \mathbf{r}}$). The total and partial scattering cross section C_s and $C_{s,m}$ with the *m*-th order could be arranged as

$$C_s = \sum_m C_{s,m} = \sum_m \frac{2\lambda}{\pi} \left| D_{i,m} \right|^2 \tag{2.7}$$

Lorentz line shape is fitted to the values of $C_{s,m}$ to derive the corresponding wavelength λ_m and p_m .

$$C_{s,m} = \frac{p_m}{[(k_0 - 2\pi/\lambda_m)^2 - Y_m^2]}$$
(2.8)

The quality factor could be evaluated as:

$$Q = 2\pi / (\lambda_m \sqrt{p_m \, k_0}) \tag{2.9}$$

Systematic examinations of self-rolled-up microtubes used for the optical resonances with versatile wall composition and smaller sizes have been conducted to identify their liquid sensing capability through the Mie scattering method. Three environment conditions have been considered including tube-in-liquid, hollow-tube-in-liquid and liquid-in-tube. As shown in Fig. 2.11 [84], the corresponding resonant wavelengths λ_r , Q factors, figures of merit QS and sensitivities S are calculated and compared with the given wall thickness Δ and diameter h, aiming at designing better optofluidic devices based on rolled-up microtubes.



Figure 2.11. The derived resonant mode and sensing capability for microtubes with refractive index $n_2 = 2$, m = 40 and different wall thickness Δ . (a) Wavelengths λ_r (b) *Q*-factors, (c) sensitivities *S* and (d) figures of merit *QS*. The symbols in (a) and (c) are approximated results obtained from waveguide approximation approaches [84]. [Figures adapted with permissions]

2.3.2 Compact on-chip self-rolled-up microtubes for lab-in-a-tube systems

Because the dimension of rolled-up structure is comparable to the microchip, the rolledup tubular microcavity can be employed as the technical platform to combine multi-functions on highly integrated devices and realise lab-in-a-tube systems [91]. A pioneer work has been shown in Fig. 2.12, which developed ultra-compact on-chip microtube arrays toward sensing and biofluid applications [92]. This microfluidic integration of several rolled-up optofluidic ring resonators on one chip is achieved by placing the patterned robust SU-8 polymeric matrix into microtubes. Due to the difference in refractive indices, different fluids passing through the microtube could be identified based on the resulting various shifting magnitudes of whispering gallery modes detected by the photoluminescence spectroscopy system. The sensitivity is measured to be up to 880 nm/refractive index units (RIU).



Figure 2.12. Ultra-compact on-chip microtubes for sensing and biofluid applications (a) SEM image shows the fabricated 3D tubular micro sensor released from a U-shape pattern. (b) Schematic illustration of the entire chip with the microchannel system of optofluidic tubular ring resonators. (c) PL spectrum for the middle part of self-rolled-up microtubes with the inset for a group of axial modes. (d) The test of signal stability and red shift for air and DI water [25]. [Figures adapted with permissions]

2.4 Self-rolled-up nanomembranes used for energy storage devices

Self-rolled-up nanomembranes have recently attracted numerous research attentions for developing novel energy storage devices, such as Lithium-ion batteries (LIBs) [93], Li-O2 batteries [94], electrode structure [30], and capacitor arrays [17], because of excellent mechanical stability and geometrical flexibility. The microfabrication based on strain induced self-rolling could scale down the battery size into microscale and thus allowing other microdevices including field effect transistors (FET), photodetectors, and logic gates to be

embedded into the micro battery and achieving the miniaturization of electronic devices [95]. The micro batteries fabricated from the self-rolled-up tubular structures could also provide the on-chip driving power and are used as an electromechanical or electrochemical device platform for electric characteristics testing [96]. Additionally, considerable efforts have been made to create the composite tubular geometry self rolling from strain engineered multiple functional materials and form hybrid organic/inorganic electronic storage devices [97]. As a demonstration of energy storage devices based on the self-rolled-up nanomembranes, Fig. 2.13 shows their applications for Li-ion batteries anode structures and hybrid organic/inorganic capacitor [93, 97]. In Fig. 2.13 (a), strained bilayer system of Si/reduced graphene oxide has been designed to roll up into many turns and form alternative sandwich nanoarchitecture with some gaps between two closed turns, aiming at facilitating electron transport and improving the tolerance of strain cracking during lithiation/delithiation cycling, which avoids large capacity loss and huge shape change of silicon anodes materials and enhances the cycling performance of LIBs. In Fig. 2.13 (b), through applying the self-rolling technique, the tubular micro-capacitors with sizes almost 2 orders of magnitude smaller than traditional ones, have been manufactured using hybrid organic/inorganic materials. They show excellent capacitances per footprint area of around 200 μ F/cm² with specific energy (~0.55 Wh/kg), much better than those metal-insulator-metal capacitors made from Al₂O₃. This advantage originates from the additional incorporations of functional organic monolayers, allowing flexible tuning of the electronic characteristics of capacities. These self-rolled-up structures coupling with embedded functional organic molecules are also promising to be extended to fabricate other microdevices like coils and transformers with desired biochemical functionalities.



Figure 2.13. Self-rolled-up nanomembranes for Li-ion batteries anode structures and hybrid organic/inorganic capacitor. (a) The cycling performance of the Si/Reduced Graphene Oxide (Si/rGO) bilayer nanomembrane electrode with the left inset showing schematic fabrication of downward rolled-up structure and right inset for the corresponding SEM image of a single rolled-up nanomembrane [93]. (b) Nanomembrane based rolled-up ultracompact capacitors (UCCap). The left region shows fabrication illustrations including the layer sequence and two cases with different consisting materials: inorganic (Case I) and hybrid organic/inorganic (Case II). The right part depicts the real SEM images of nanomembrane-based UCCaps [97]. [Figures adapted with permissions]

2.4.1 Rolled-up nanomembranes used for magnetic tubular architectures

It has been revealed that, experimentally and theoretically, the curved surface of magnet

would result in polarity-chirality coupling [98], magneto chiral properties [99], magnetization

patterning [56] and anisotropic magneto static interaction [58]. The successful fabrications of various self-rolled-up magnetic shell structures with controllable curvature have shown great potentials for the investigations of those curvature-driven modifications of physical properties to magnetic nano-objects [99-101]. Additionally, those self-rolled-up magnetic structures have also been explored for the related magnetic applications, such as, flow detection of magnetic objects [102], magneto-encephalography devices [103], and ferromagnetic microtube ring resonators [57] through depositing an additional magnetic layer. Currently, more efforts are devoted to optimizing the initial heterostructures with magnetic material incorporated to achieve smaller diameters down to several nanometers and thus maximizing the curvature effects. Fig. 2.14 gives the application of spin-wave ferromagnetic microtube ring resonators made from strained semiconductor bilayer of InGaAs/GaAs with a ferromagnetic metal layer Ni₈₀Fe₂₀ [57]. The spin-wave spectrum has been measured through microwave absorption spectroscopy, and the results indicate that zero magnetic field mode spectrum could be effectively tailored the diameter and total wall thickness of the microtube. Moreover, this tubular ferromagnetic structure can produce multiple separated whispering-gallery modes over a wide magnetic field (several GHz).



Figure 2.14. Spin-wave ring resonators made from rolled-up ferromagnetic permalloy microtube (RUPT) based on $Ni_{80}Fe_{20}/GaAs/In_{20}Ga_{80}As$ (a) illustrations for the layer sequence of strain engineered $Ni_{80}Fe_{20}/GaAs/In_{20}Ga_{80}As$. (b) and (c): the schematic diagram and corresponding SEM image of RUPT placed along the signal line (S) between two ground lines (G). The left inset shows the cross-section window of microtube with 3.5 rolling turns and the following microwave absorption measurements are conduced by a coplanar waveguide [57]. [Figures adapted with permissions]

2.4.2 Self-rolling method extended for the application of polymer structures

Recently, the concept of strain-induced self-rolling, initially developed for fabricating semiconductor nanomembranes, has been extended to other material systems, particularly for the polymer-based self-folding structures [1, 104]. The main advantages of polymer materials lie in their sensitivities to various external signals (like temperature and pH) and considerable tolerance of elastic deformation without considering the limitation of small deformations and the linear elastic behaviors of semiconductor materials [43, 105]. Additionally, the spatial

arrangement of strain engineered polymers could be produced by the biomimetic 4D printing with designed anisotropic strain gradient when responding to external stimuli [106]. As examples, polymer-based origami has been introduced as the new applications of self-rolling techniques, which has made possible the creation of an enormous amount of new designs, and enables multi-stability that allows reprogrammable configurations [107, 108]. Moreover, appropriate software package based on origami mathematics has been available and ready to solve the inverse origami problems and design significantly complex 3D origami geometry [109]. Some particularly interesting results are shown in Fig. 2.15, where the cubic box, dodecahedron and Miura-ori folding pattern have been created through the connected polymer panels driven by the temperature induced rolling elements [107]. Although the fundamental geometry and elastic theory of origami-based metamaterials have been addressed [110, 111], several issues need to be further discussed and improved with respect to the self-rolling behaviors of connected elements in those origami patterns, especially for the effects of geometrical and mechanical parameters including the thickness, elastic constants, temperature induced strain gradient, and the controlled actuation sequence [112]. Also, the polymers are not suitable for the structural and actuation applications due to the low modulus characteristics.



Figure 2.15. Printing self-rolling elements activated by the temperature (enclosed in the white rectangles) connecting the panels for creating origami structures. (a) Six connected square panels self rolling into a cubic box after temperature change. (b) Connected panels self-rolled into a dodecahedron (c) The Miura-ori folding pattern fabricated by printing two types of elements with opposite rolling directions upon activation of temperature [107]. [Figures adapted with permissions]

2.5 References

[1] Y.H. Zhang, F. Zhang, Z. Yan, Q. Ma, X.L. Li, Y.G. Huang, J.A. Rogers, Printing, Folding and Assembly Methods for Forming 3d Mesostructures in Advanced Materials (Vol 2, Pg 17019, 2017), Nat Rev Mater, 2 (2017).

[2] L.Z. Xu, T.C. Shyu, N.A. Kotov, Origami and Kirigami Nanocomposites, Acs Nano, 11 (2017) 7587-7599.

[3] H.J. Bae, S. Bae, J. Yoon, C. Park, K. Kim, S. Kwon, W. Park, Self-Organization of Maze-Like Structures Via Guided Wrinkling, Sci Adv, 3 (2017).

[4] Y. Wang, Z.W. Li, J.L. Xiao, Stretchable Thin Film Materials: Fabrication, Application, and Mechanics, J Electron Packaging, 138 (2016).

[5] S. Xu, Z. Yan, K.I. Jang, W. Huang, H.R. Fu, J. Kim, Z. Wei, M. Flavin, J. McCracken, R. Wang, A. Badea, Y. Liu, D.Q. Xiao, G.Y. Zhou, J. Lee, H.U. Chung, H.Y. Cheng, W. Ren, A. Banks, X.L. Li, U. Paik, R.G. Nuzzo, Y.G. Huang, Y.H. Zhang, J.A. Rogers, Assembly of Micro/Nanomaterials into Complex, Three-Dimensional Architectures by Compressive Buckling, Science, 347 (2015) 154-159.

[6] Z. Yan, F. Zhang, F. Liu, M.D. Han, D.P. Ou, Y.H. Liu, Q. Lin, X.L. Guo, H.R. Fu, Z.Q. Xie, M.Y. Gao, Y.M. Huang, J. Kim, Y.T. Qiu, K.W. Nan, J. Kim, P. Gutruf, H.Y. Luo, A. Zhao, K.C. Hwang, Y.G. Huang, Y.H. Zhang, J.A. Rogers, Mechanical Assembly of Complex, 3d Mesostructures from Releasable Multilayers of Advanced Materials, Sci Adv, 2 (2016).

[7] D. Mijatovic, J.C.T. Eijkel, A. van den Berg, Technologies for Nanofluidic Systems: Top-Down Vs. Bottom-up - a Review, Lab Chip, 5 (2005) 492-500.

[8] H.R. Kwag, J.H. Cho, S.Y. Park, J. Park, D.H. Gracias, Self-Folding Nanostructures with Imprint Patterned Surfaces (Snips), Faraday Discuss, 191 (2016) 61-71.

[9] R.G. Hobbs, N. Petkov, J.D. Holmes, Semiconductor Nanowire Fabrication by Bottom-up and Top-Down Paradigms, Chem Mater, 24 (2012) 1975-1991.

[10] T. Sakanoue, M. Mizukami, S. Oku, Y. Yoshimura, M. Abiko, S. Tokito, Fluorosurfactant-Assisted Photolithography for Patterning of Perfluoropolymers and Solution-Processed Organic Semiconductors for Printed Displays, Appl Phys Express, 7 (2014).

[11] R. Garcia, A.W. Knoll, E. Riedo, Advanced Scanning Probe Lithography, Nature Nanotechnology, 9 (2014) 577-587.

[12] J.K.Y. Ong, D. Moore, J. Kane, R.F. Saraf, Negative Printing by Soft Lithography, Acs Appl Mater Inter, 6 (2014) 14278-14285.

[13] H. Schift, Nanoimprint Lithography: 2d or Not 2d? A Review, Applied Physics A, 121 (2015) 415-435.

[14] C.E. Morosanu, G. Siddall, Thin Films by Chemical Vapour Deposition, Elsevier Science, 2016.

[15] J.Y. Tsao, Materials Fundamentals of Molecular Beam Epitaxy, Elsevier Science, 2012.

[16] K. Gregorczyk, M. Knez, Hybrid Nanomaterials through Molecular and Atomic Layer Deposition: Top Down, Bottom up, and in-between Approaches to New Materials, Prog Mater Sci, 75 (2016) 1-37.

[17] R. Sharma, C.C.B. Bufon, D. Grimm, R. Sommer, A. Wollatz, J. Schadewald, D.J. Thurmer, P.F. Siles, M. Bauer, O.G. Schmidt, Large-Area Rolled-up Nanomembrane Capacitor Arrays for Electrostatic Energy Storage, Adv Energy Mater, 4 (2014).

[18] Y.F. Mei, G.S. Huang, A.A. Solovev, E.B. Urena, I. Moench, F. Ding, T. Reindl, R.K.Y. Fu, P.K. Chu, O.G. Schmidt, Versatile Approach for Integrative and Functionalized Tubes by Strain Engineering of Nanomembranes on Polymers, Adv Mater, 20 (2008) 4085-4090.

[19] D. Grimm, R.B. Wilson, B. Teshome, S. Gorantla, M.H. Rummeli, T. Bublat, E. Zallo, G.D. Li, D.G. Cahill, O.G. Schmidt, Thermal Conductivity of Mechanically Joined Semiconducting/Metal Nanomembrane Superlattices, Nano Lett, 14 (2014) 2387-2393.

[20] J.W. Deng, X.Y. Lu, L.X. Liu, L. Zhang, O.G. Schmidt, Introducing Rolled-up Nanotechnology for Advanced Energy Storage Devices, Adv Energy Mater, 6 (2016).

[21] I.S. Chun, X.L. Li, Controlled Assembly and Dispersion of Strain-Induced Ingaas/Gaas Nanotubes, Ieee T Nanotechnol, 7 (2008) 493-495.

[22] F. Li, Z.T. Mi, Optically Pumped Rolled-up Ingaas/Gaas Quantum Dot Microtube Lasers, Optics Express, 17 (2009) 19933-19939.

[23] D. Grimm, C.C.B. Bufon, C. Deneke, P. Atkinson, D.J. Thurmer, F. Schaffel, S. Gorantla, A. Bachmatiuk, O.G. Schmidt, Rolled-up Nanomembranes as Compact 3d Architectures for Field Effect Transistors and Fluidic Sensing Applications, Nano Lett, 13 (2013) 213-218.

[24] P. Froeter, Y. Huang, O.V. Cangellaris, W. Huang, E.W. Dent, M.U. Gillette, J.C. Williams, X.L. Li, Toward Intelligent Synthetic Neural Circuits: Directing and Accelerating Neuron Cell Growth by Self-Rolled-up Silicon Nitride Microtube Array, Acs Nano, 8 (2014) 11108-11117.

[25] S. Sanchez, Lab-in-a-Tube Systems as Ultra-Compact Devices, Lab Chip, 15 (2015) 610-613.

[26] L. Persano, A. Camposeo, D. Pisignano, Integrated Bottom-up and Top-Down Soft Lithographies and Microfabrication Approaches to Multifunctional Polymers, J Mater Chem C, 1 (2013) 7663-7680.

[27] J. Wu, Y.Y. Liu, Y.Y. Guo, S.L. Feng, B.H. Zou, H. Mao, C.H. Yu, D.B. Tian, W. Huang, F.W. Huo, Centimeter-Scale Subwavelength Photolithography Using Metal-Coated Elastomeric Photomasks with Modulated Light Intensity at the Oblique Sidewalls, Langmuir, 31 (2015) 5005-5013.

[28] S.F. Huang, H.F. Zhang, Z.L. Wu, D.Z. Kong, D.D. Lin, Y.L. Fan, X.J. Yang, Z.Y. Zhong, S.H. Huang, Z.M. Jiang, C.W. Cheng, Large-Area Ordered P-Type Si Nanowire Arrays as Photocathode for Highly Efficient Photoelectrochemical Hydrogen Generation, Acs Appl Mater Inter, 6 (2014) 12111-12118.

[29] S.Z. Huang, L. Zhang, X.Y. Lu, L.F. Liu, L.X. Liu, X.L. Sun, Y. Yin, S. Oswald, Z.Y. Zou, F. Ding, O.G. Schmidt, Tunable Pseudocapacitance in 3d Tio2-Delta Nanomembranes Enabling Superior Lithium Storage Performance, Acs Nano, 11 (2017) 821-830.

[30] L. Zhang, J.W. Deng, L.F. Liu, W.P. Si, S. Oswald, L.X. Xi, M. Kundu, G.Z. Ma, T. Gemming, S. Baunack, F. Ding, C.L. Yan, O.G. Schmidt, Hierarchically Designed Siox/Sioy Bilayer Nanomembranes as Stable Anodes for Lithium Ion Batteries, Adv Mater, 26 (2014) 4527-4532.

[31] C.L. Yan, W. Xi, W.P. Si, J.W. Deng, O.G. Schmidt, Highly Conductive and Strain-Released Hybrid Multilayer Ge/Ti Nanomembranes with Enhanced Lithium-Ion-Storage Capability, Adv Mater, 25 (2013) 539-544.

[32] H. Wang, M. Pumera, Fabrication of Micro/Nanoscale Motors, Chem Rev, 115 (2015) 8704-8735.

[33] Y.F. Mei, A.A. Solovev, S. Sanchez, O.G. Schmidt, Rolled-up Nanotech on Polymers: From Basic Perception to Self-Propelled Catalytic Microengines, Chem Soc Rev, 40 (2011) 2109-2119.

[34] J. Wang, T.R. Zhan, G.S. Huang, P.K. Chu, Y.F. Mei, Optical Microcavities with Tubular Geometry: Properties and Applications, Laser Photonics Rev, 8 (2014) 521-547.

[35] S.L. Li, L.B. Ma, H.L. Zhen, M.R. Jorgensen, S. Kiravittaya, O.G. Schmidt, Dynamic Axial Mode Tuning in a Rolled-up Optical Microcavity, Appl Phys Lett, 101 (2012).

[36] S.W. Tang, Y.F. Fang, Z.W. Liu, L. Zhou, Y.F. Mei, Tubular Optical Microcavities of Indefinite Medium for Sensitive Liquid Refractometers, Lab Chip, 16 (2016) 182-187.

[37] O.G. Schmidt, K. Eberl, Nanotechnology - Thin Solid Films Roll up into Nanotubes, Nature, 410 (2001) 168-168.

[38] V.Y. Prinz, V.A. Seleznev, A.K. Gutakovsky, A.V. Chehovskiy, V.V. Preobrazhenskii, M.A. Putyato, T.A. Gavrilova, Free-Standing and Overgrown Ingaas/Gaas Nanotubes, Nanohelices and Their Arrays, Physica E, 6 (2000) 828-831.

[39] I.S. Chun, A. Challa, B. Derickson, K.J. Hsia, X.L. Li, Geometry Effect on the Strain-Induced Self-Rolling of Semiconductor Membranes, Nano Lett, 10 (2010) 3927-3932.

[40] R. Arayanarakool, A.K. Meyer, L. Helbig, S. Sanchez, O.G. Schmidt, Tailoring Three-Dimensional Architectures by Rolled-up Nanotechnology for Mimicking Microvasculatures, Lab Chip, 15 (2015) 2981-2989.

[41] D.J. Bell, L.X. Dong, B.J. Nelson, M. Golling, L. Zhang, D. Grutzmacher, Fabrication and Characterization of Three-Dimensional Ingaas/Gaas Nanosprings, Nano Lett, 6 (2006) 725-729.
[42] T. Ma, H.S. Liang, G. Chen, B. Poon, H.Q. Jiang, H.B. Yu, Micro-Strain Sensing Using Wrinkled Stiff Thin Films on Soft Substrates as Tunable Optical Grating, Optics Express, 21 (2013) 11994-12001.

[43] D.H. Gracias, Stimuli Responsive Self-Folding Using Thin Polymer Films, Curr Opin Chem Eng, 2 (2013) 112-119.

[44] L. Ionov, Hydrogel-Based Actuators: Possibilities and Limitations, Materials Today, 17 (2014) 494-503.

[45] I.S. Chun, V.B. Verma, V.C. Elarde, S.W. Kim, J.M. Zuo, J.J. Coleman, X. Li, Ingaas/Gaas 3d Architecture Formation by Strain-Induced Self-Rolling with Lithographically Defined Rectangular Stripe Arrays, J Cryst Growth, 310 (2008) 2353-2358.

[46] L. Zhang, E. Ruh, D. Grutzmacher, L.X. Dong, D.J. Bell, B.J. Nelson, C. Schonenberger, Anomalous Coiling of Sige/Si and Sige/Si/Cr Helical Nanobelts, Nano Lett, 6 (2006) 1311-1317.

[47] P. Cendula, S. Kiravittaya, I. Monch, J. Schumann, O.G. Schmidt, Directional Roll-up of Nanomembranes Mediated by Wrinkling, Nano Lett, 11 (2011) 236-240.

[48] W. Huang, S. Koric, X. Yu, K.J. Hsia, X.L. Li, Precision Structural Engineering of Self-Rolled-up 3d Nanomembranes Guided by Transient Quasi-Static Fem Modeling, Nano Lett, 14 (2014) 6293-6297.

[49] Z. Chen, G.S. Huang, I. Trase, X.M. Han, Y.F. Mei, Mechanical Self-Assembly of a Strain-Engineered Flexible Layer: Wrinkling, Rolling, and Twisting, Phys Rev Appl, 5 (2016).

[50] T. Huang, Z.Q. Liu, G.S. Huang, R. Liu, Y.F. Mei, Grating-Structured Metallic Microsprings, Nanoscale, 6 (2014) 9428-9435.

[51] B.C. Li, Q. Wang, E.Y. Wang, Z.G. Jia, Y.C. Yan, Z.Q. Bian, X.M. Ren, S.W. Cai, Y.Q. Huang, Dependence of Doubly Curved Regions on Drying Method in the Fabrication of Long-Side Rolled-up Iii-V Microtubes, Appl Phys Lett, 103 (2013).

[52] S. Alben, B. Balakrisnan, E. Smela, Edge Effects Determine the Direction of Bilayer Bending, Nano Lett, 11 (2011) 2280-2285.

[53] L. Zhang, E. Deckhardt, A. Weber, C. Schonenberger, D. Grutzmacher, Controllable Fabrication of Sige/Si and Sige/Si/Cr Helical Nanobelts, Nanotechnology, 16 (2005) 655-663.

[54] D.H. Kim, J.H. Ahn, W.M. Choi, H.S. Kim, T.H. Kim, J.Z. Song, Y.G.Y. Huang, Z.J. Liu, C. Lu, J.A. Rogers, Stretchable and Foldable Silicon Integrated Circuits, Science, 320 (2008) 507-511.

[55] Y. Gaididei, V.P. Kravchuk, D.D. Sheka, Curvature Effects in Thin Magnetic Shells, Phys Rev Lett, 112 (2014).

[56] O.V. Pylypovskyi, V.P. Kravchuk, D.D. Sheka, D. Makarov, O.G. Schmidt, Y. Gaididei, Coupling of Chiralities in Spin and Physical Spaces: The Mobius Ring as a Case Study, Phys Rev Lett, 114 (2015).

[57] F. Balhorn, S. Mansfeld, A. Krohn, J. Topp, W. Hansen, D. Heitmann, S. Mendach, Spin-Wave Interference in Three-Dimensional Rolled-up Ferromagnetic Microtubes, Phys Rev Lett, 104 (2010). [58] R. Streubel, D.J. Thurmer, D. Makarov, F. Kronast, T. Kosub, V. Kravchuk, D.D. Sheka, Y. Gaididei, R. Schafer, O.G. Schmidt, Magnetically Capped Rolled-up Nanomembranes, Nano Lett, 12 (2012) 3961-3966.

[59] B. Huang, K.H. Jin, B. Cui, F. Zhai, J.W. Mei, F. Liu, Bending Strain Engineering in Quantum Spin Hall System for Controlling Spin Currents, Nat Commun, 8 (2017).

[60] L. Ionov, 3d Microfabrication Using Stimuli-Responsive Self-Folding Polymer Films, Polym Rev, 53 (2013) 92-107.

[61] E. Efrati, E. Sharon, R. Kupferman, Elastic Theory of Unconstrained Non-Euclidean Plates, J Mech Phys Solids, 57 (2009) 762-775.

[62] S. Timoshenko, S. Woinowsky-Krieger, Theory of Plates and Shells, McGraw-Hill, 1959.
[63] L. Zhang, S.V. Golod, E. Deckardt, V. Prinz, D. Grutzmacher, Free-Standing Si/Sige Micro- and Nano-Objects, Physica E, 23 (2004) 280-284.

[64] C. Deneke, C. Muller, N.Y. Jin-Phillipp, O.G. Schmidt, Diameter Scalability of Rolled-up in(Ga)as/Gaas Nanotubes, Semicond Sci Tech, 17 (2002) 1278-1281.

[65] J. Zang, F. Liu, Modified Timoshenko Formula for Bending of Ultrathin Strained Bilayer Films, Appl Phys Lett, 92 (2008).

[66] M. Grundmann, Nanoscroll Formation from Strained Layer Heterostructures, Appl Phys Lett, 83 (2003) 2444-2446.

[67] O. Schumacher, S. Mendach, H. Welsch, A. Schramm, C. Heyn, W. Hansen, Lithographically Defined Metal-Semiconductor-Hybrid Nanoscrolls, Appl Phys Lett, 86 (2005).
[68] J. Zang, M.H. Huang, F. Liu, Mechanism for Nanotube Formation from Self-Bending Nanofilms Driven by Atomic-Scale Surface-Stress Imbalance, Phys Rev Lett, 98 (2007).

[69] M.H. Huang, F. Cavallo, F. Liu, M.G. Lagally, Nanomechanical Architecture of Semiconductor Nanomembranes, Nanoscale, 3 (2011) 96-120.

[70] X.L. Li, Strain Induced Semiconductor Nanotubes: From Formation Process to Device Applications, J Phys D Appl Phys, 41 (2008).

[71] Y.F. Mei, D.J. Thurmer, C. Deneke, S. Kiravittaya, Y.F. Chen, A. Dadgar, F. Bertram, B. Bastek, A. Krost, J. Christen, T. Reindl, M. Stoffel, E. Coric, O.G. Schmidt, Fabrication, Self-Assembly, and Properties of Ultrathin Aln/Gan Porous Crystalline Nanomembranes: Tubes, Spirals, and Curved Sheets, Acs Nano, 3 (2009) 1663-1668.

[72] S.Z. Zhu, T. Li, Hydrogenation-Assisted Graphene Origami and Its Application in Programmable Molecular Mass Uptake, Storage, and Release, Acs Nano, 8 (2014) 2864-2872.
[73] H.S. Lee, M. Cho, B.J. Lee, Size Dependence in Self-Bending Mechanism of Ge/Si Bilayer Nanofilms, J Appl Phys, 109 (2011).

[74] D. Joung, A. Nemilentsau, K. Agarwal, C.H. Dai, C. Liu, Q. Su, J. Li, T. Low, S.J. Koester, J.H. Cho, Self-Assembled Three-Dimensional Graphene-Based Polyhedrons Inducing Volumetric Light Confinement, Nano Lett, 17 (2017) 1987-1994.

[75] X.L. Li, Self-Rolled-up Microtube Ring Resonators: A Review of Geometrical and Resonant Properties, Adv Opt Photonics, 3 (2011) 366-387.

[76] G.S. Huang, Y.F. Mei, Electromagnetic Wave Propagation in a Rolled-up Tubular Microcavity, J Mater Chem C, 5 (2017) 2758-2770.

[77] T.R. Zhan, C. Xu, F.Y. Zhao, Z.Q. Xiong, X.H. Hu, G.S. Huang, Y.F. Mei, J. Zi, Optical Resonances in Tubular Microcavities with Subwavelength Wall Thicknesses, Appl Phys Lett, 99 (2011).

[78] G.S. Huang, V.A.B. Quinones, F. Ding, S. Kiravittaya, Y.F. Mei, O.G. Schmidt, Rolledup Optical Microcavities with Subwavelength Wall Thicknesses for Enhanced Liquid Sensing Applications, Acs Nano, 4 (2010) 3123-3130.

[79] X.Y. Lin, Y.F. Fang, L.J. Zhu, J. Zhang, G.S. Huang, J. Wang, Y.F. Mei, Self-Rolling of Oxide Nanomembranes and Resonance Coupling in Tubular Optical Microcavity, Adv Opt Mater, 4 (2016) 936-942.

[80] X. Yu, L.L. Goddard, X.L. Li, X.G. Chen, Enhanced Axial Confinement in a Monolithically Integrated Self-Rolled-up Sinx Vertical Microring Photonic Coupler, Appl Phys Lett, 109 (2016).

[81] S. Bottner, M.R. Jorgensen, O.G. Schmidt, Rolled-up Nanotechnology: 3d Photonic Materials by Design, Scripta Mater, 122 (2016) 119-124.

[82] X. Yu, E. Arbabi, L.L. Goddard, X.L. Li, X.G. Chen, Monolithically Integrated Self-Rolled-up Microtube-Based Vertical Coupler for Three-Dimensional Photonic Integration, Appl Phys Lett, 107 (2015).

[83] Y. Yin, Y. Chen, E.S.G. Naz, X.Y. Lu, S.L. Li, V. Engemaier, L.B. Ma, O.G. Schmidt, Silver Nanocap Enabled Conversion and Tuning of Hybrid Photon-Plasmon Modes in Microtubular Cavities, Acs Photonics, 4 (2017) 736-740.

[84] F.Y. Zhao, T.R. Zhan, G.S. Huang, Y.F. Mei, X.H. Hu, Liquid Sensing Capability of Rolled-up Tubular Optical Microcavities: A Theoretical Study, Lab Chip, 12 (2012) 3798-3802.
[85] J. Wang, T.R. Zhan, G.S. Huang, X.G. Cui, X.H. Hu, Y.F. Mei, Tubular Oxide Microcavity with High-Index-Contrast Walls: Mie Scattering Theory and 3d Confinement of Resonant Modes, Optics Express, 20 (2012) 18555-18567.

[86] T. Kipp, H. Welsch, C. Strelow, C. Heyn, D. Heitmann, Optical Modes in Semiconductor Microtube Ring Resonators, Phys Rev Lett, 96 (2006).

[87] J. Zhang, J. Zhong, Y.F. Fang, J. Wang, G.S. Huang, X.G. Cui, Y.F. Mei, Roll up Polymer/Oxide/Polymer Nanomembranes as a Hybrid Optical Microcavity for Humidity Sensing, Nanoscale, 6 (2014) 13646-13650.

[88] M. Hosoda, T. Shigaki, Degeneracy Breaking of Optical Resonance Modes in Rolled-up Spiral Microtubes, Appl Phys Lett, 90 (2007).

[89] a. Bernardi, S. Kiravittaya, a. Rastelli, R. Songmuang, D.J. Thurmer, M. Benyoucef, O.G. Schmidt, On-Chip Si/Siox Microtube Refractometer, Appl Phys Lett, 93 (2008).

[90] S.M. Harazim, V.A.B. Quinones, S. Kiravittaya, S. Sanchez, O.G. Schmidt, Lab-in-a-Tube: On-Chip Integration of Glass Optofluidic Ring Resonators for Label-Free Sensing Applications, Lab Chip, 12 (2012) 2649-2655.

[91] S. Ivaro, G. Marn, S. Vincent, I.G. Marn, W.V. Hoeve, P. Garca-snchez, N. Convine, A. Rosser-james, M. Tyler, K. Bandoo, L. Warncke, A. Lee, V. Vogel, Lab-in-a-Tube Systems as Ultra-Compact Devices, Lab Chip, 15 (2015) 4491--4498.

[92] S.M.a. Harazim, Lab-in-a-Tube: On-Chip Integration of Glass Optofluidic Ring Resonators for Label-Free Sensing Applications, Lab Chip, 12 (2012) 2649.

[93] X.H. Liu, J. Zhang, W.P. Si, L.X. Xi, B. Eichler, C.L. Yan, O.G. Schmidt, Sandwich Nano Architecture of Si/Reduced Graphene Oxide Bilayer Nanomembranes for Li-Ion Batteries with Long Cycle Life, Acs Nano, 9 (2015) 1198-1205.

[94] X.Y. Lu, J.W. Deng, W.P. Si, X.L. Sun, X.H. Liu, B. Liu, L.F. Liu, S. Oswald, S. Baunack, H.J. Grafe, C.L. Yan, O.G. Schmidt, High-Performance Li-O-2 Batteries with Trilayered Pd/Mnox/Pd Nanomembranes, Adv Sci, 2 (2015).

[95] X.F. Wang, Y. Chen, O.G. Schmidt, C.L. Yan, Engineered Nanomembranes for Smart Energy Storage Devices, Chem Soc Rev, 45 (2016) 1308-1330.

[96] W.P. Si, I. Monch, C.L. Yan, J.W. Deng, S.L. Li, G.G. Lin, L.Y. Han, Y.F. Mei, O.G. Schmidt, A Single Rolled-up Si Tube Battery for the Study of Electrochemical Kinetics, Electrical Conductivity, and Structural Integrity, Adv Mater, 26 (2014) 7973-7978.

[97] C.C.B. Bufon, J.D.C. Gonzalez, D.J. Thurmer, D. Grimm, M. Bauer, O.G. Schmidt, Self-Assembled Ultra-Compact Energy Storage Elements Based on Hybrid Nanomembranes, Nano Lett, 10 (2010) 2506-2510.

[98] V.P. Kravchuk, D.D. Sheka, R. Streubel, D. Makarov, O.G. Schmidt, Y. Gaididei, Out-of-Surface Vortices in Spherical Shells, Phys Rev B, 85 (2012).

[99] R. Hertel, Curvature-Induced Magnetochirality, Spin-Singapore, 3 (2013).

[100] R. Streubel, P. Fischer, F. Kronast, V.P. Kravchuk, D.D. Sheka, Y. Gaididei, O.G. Schmidt, D. Makarov, Magnetism in Curved Geometries, J Phys D Appl Phys, 49 (2016).

[101] X.W. Zhou, R.E. Jones, J. Gruber, Molecular Dynamics Simulations of Substitutional Diffusion, Comp Mater Sci, 128 (2017) 331-336.

[102] I. Monch, D. Makarov, R. Koseva, L. Baraban, D. Karnaushenko, C. Kaiser, K.F. Arndt, O.G. Schmidt, Rolled-up Magnetic Sensor: Nanomembrane Architecture for in-Flow Detection of Magnetic Objects, Acs Nano, 5 (2011) 7436-7442.

[103] D. Karnaushenko, D.D. Karnaushenko, D. Makarov, S. Baunack, R. Schafer, O.G. Schmidt, Self-Assembled on-Chip-Integrated Giant Magneto-Impedance Sensorics, Adv Mater, 27 (2015) 6582-6589.

[104] A. Azam, K.E. Laflin, M. Jamal, R. Fernandes, D.H. Gracias, Self-Folding Micropatterned Polymeric Containers, Biomed Microdevices, 13 (2011) 51-58.

[105] Q. Ge, A.H. Sakhaei, H. Lee, C.K. Dunn, N.X. Fang, M.L. Dunn, Multimaterial 4d Printing with Tailorable Shape Memory Polymers, Sci Rep-Uk, 6 (2016).

[106] A.S. Gladman, E.A. Matsumoto, R.G. Nuzzo, L. Mahadevan, J.A. Lewis, Biomimetic 4d Printing, Nat Mater, 15 (2016) 413-418.

[107] T. van Manen, S. Janbaz, A.A. Zadpoor, Programming 2d/3d Shape-Shifting with Hobbyist 3d Printers, Materials Horizons, (2017).

[108] J.H. Na, A.A. Evans, J. Bae, M.C. Chiappelli, C.D. Santangelo, R.J. Lang, T.C. Hull, R.C. Hayward, Programming Reversibly Self-Folding Origami with Micropatterned Photo-Crosslinkable Polymer Trilayers, Adv Mater, 27 (2015) 79-85.

[109] J. Rogers, Y.G. Huang, O.G. Schmidt, D.H. Gracias, Origami Mems and Nems, Mrs Bull, 41 (2016) 123-129.

[110] V. Brunck, F. Lechenault, A. Reid, M. Adda-Bedia, Elastic Theory of Origami-Based Metamaterials, Phys Rev E, 93 (2016).

[111] S. Waitukaitis, R. Menaut, B.G.G. Chen, M. van Hecke, Origami Multistability: From Single Vertices to Metasheets, Phys Rev Lett, 114 (2015).

[112] N. An, M.E. Li, J.X. Zhou, Predicting Origami-Inspired Programmable Self-Folding of Hydrogel Trilayers, Smart Mater Struct, 25 (2016).

Chapter 3: Research Methodology

The precise design and manipulation of the rolled-up geometries of strain engineered nanomembrane fundamentally requires deep understanding of underlying self-rollup mechanics and dynamics evolution mechanism. In this research, the self-rollup behaviors in continuum regimes were studied through Finite element (FE) analysis combined with continuum mechanics theory, which helps to deal with the geometrically complex strained multilayered nanomembranes with material anisotropy and predefined topological patterns. However, those approaches fail to account for the role of lattice defects (e.g., dislocations) in modifying the strain gradient of nanomembranes, where the key information about the exact atomic arrangement of rolled up structures needs to be further considered. Therefore, molecular dynamics (MD) simulations have been performed to examine the self-rolling mechanics providing atomistic details of how self rolling systems operate. The combination of those methods would allow the rational design and construct of flexible rollup geometries for various applications with less cost and development cycles. This chapter will introduce relevant principle and concepts of modeling and simulation methods used in this thesis and describe possible limitations.

3.1 Finite element (FE) analysis using ABAQUS

Most practical design of rollup configurations obtained from strain-induced nanomembrane involve predefined region with a complicated 3D structure [22], and need to consider inherently material behaviors such as the contact with surface functional patterns [23], large local curvature and volume changes [6] as well as nonlinear elastic effects [24]. These problems are beyond the reach of the traditional analytical techniques and could only be treated using numerical simulations, and the Finite element (FE) analysis is probably the most versatile

and widely used numerical technique developed to deal with those issues, which has a strong advantage on the investigation of mechanics of deformable solids of irregular geometry coupled with the problems of heat and mass transfer, fluid flow and electromagnetic field effects [25]. It mainly involves subdividing the complicated 3D geometry of solid into smaller and distinct regions referred to finite elements and then, discrete points (known as nodal points) are assigned elaborately inside of these finite elements to construct the interpolating functions and related partial differential equations that governs the targeted problems, which could then be approximately solved using numerical techniques, and assembled into a larger system of equations that describes the entire problem. The approximate solutions to corresponding partial differential equations with the targeted unknowns, such as the displacements and rotations of each divided element, are generally determined employing the principle of virtual work based on the calculus of variations through minimizing associated error functions [26]. This section would give a brief introduction of the FE simulations implemented by the ABAQUS Unified FE software [27] for predicting strain-induced self rolling behaviors of strained nanomembranes.

3.1.1 ABAQUS/CAE simulations

Through the integration of open-source programming language Python for scripting and customization [28], the ABAQUS/CAE (Computer Aided Engineering) with version 6.13-1 used in our study, provides an interactive graphic interface for the high efficient FE analysis of mechanical components and assemblies, particularly for nonlinear engineering problems. It consists of a series of logical modules that describe different aspect of the modeling procedures, for example, creating the geometry, defining the mesh and assigning material properties and loading. In our simulations, the module of composite layups develop for shell geometry is used to simulate the strained bilayer or multilayer structures with or without predefined surface functional patterns [29]. The shell element labelled S4R is employed to analyze the roll-up

process numerically of strained nanomembranes. In ABAQUS documentation, S4R belongs one of general-purpose conventional shell elements, representing the 4-node, quadrilateral, stress/displacement shell element with reduced integration and a large-strain formulation, allowing transverse shear deformation [30]. Different thermal expansion coefficients are assigned to each layer to achieve the mismatch strain.

3.1.2 Comparative Selection of FE Analysis Modules

Two main analysis modules, namely ABAQUS/Standard and ABAQUS/Explicit, are used in our simulations. ABAQUS/Standard is designed for static analysis by solving a system of equations implicitly at each solution increment for a variety of linear and nonlinear problems involving static, low-speed dynamic, thermal, and electrical response of components, where highly reliable and precise solutions to stress and displacement are of great importance [31]. Thus, mechanical equilibrium is required to be achieved through each iterative procedure of ABAQUS/Standard. Comparatively speaking, ABAQUS/Explicit is a special-purpose analysis module based on the explicit dynamic finite element formulation [32]. The explicit time integration method is used to consider the kinematic conditions at one increment and to calculate the kinematic conditions at the next increment. It is suitable for modeling brief and transient dynamic events, such as impact problems with the requirement of small time increments. In this thesis, the ABAQUS/Standard module was employed to investigate the rollup process under controlled release and measure the final rolling curvature. However, it is unpractical and beyond the capability of ABAQUS/Standard module to analyze a nonlinear problem with many degrees of freedoms during a dynamic process, especially for the self rollup competitions and potential possible multiple 3D rollup configurations [33]. That is mainly attributed to the numerical divergent issues for each iteration calculation of ABAQUS/Standard, where mechanical equilibrium is required to be checked for the internal structure forces with the externally applied loads during the solution process of a large set of linear equations. On

the other hand, ABAQUS/Explicit updates the structure geometry changes without considering the iterating to enforce mechanical equilibrium, which solves the divergence problem over the implicit method but many smaller increments are required for good accuracy and time consuming [34]. Therefore, ABAQUS explicit module has the advantage for dynamic simulations of the competition between different roll-up directions after simultaneous release of strained nanomembranes.

3.2 Fundamental Continuum Theory

The fundamental continuum theories used for the quantitative analysis of self-rolling of strained nanomembranes included the von Karman shell theory for the rollup of flat plates and membranes [35], and related dislocation theory to evaluate the role of misfit dislocations on the self-rolling up [36]. This section will present a brief introduction of continuum theories pertaining to the thesis work.

3.2.1 The von Karman shell theory for flat plates and membranes

The von Karman shell theory specifically describes the mechanics of flat plates and membranes with small in-plane and large transverse deflections, which is mainly focused on the deformation of thin shell subjected to substantial loading parallel to the plane of the shell with large out-of-plane displacement [37].

The following assumptions have been made for the von Karman shell theory:

1. The displacement field within the shell conforms to the Kirchhoff shell theory [38], which states that in the shell straight lines normal to the underformed mid-surface remain straight and normal after deformation, and the thickness of the plate does not change after a deformation.

It is assumed that the in-plane loadings are much larger than transverse ones, and only nonlinear terms in the governing equations involving in-plane loadings are kept.
 The basic governing equations have been summarized as following with Greek subscripts being 1 or 2 and index 3 labelling out of plane direction [35]:

1. The in-plane strain tensor is approximated by
$$\gamma_{\alpha\beta} = \frac{1}{2} \left(\frac{\partial u_{\alpha}}{\partial x_{\beta}} + \frac{\partial u_{\beta}}{\partial x_{\alpha}} + \frac{\partial u_{3}}{\partial x_{\beta}} \cdot \frac{\partial u_{3}}{\partial x_{\alpha}} \right)$$

2. The curvature change tensor has components $\Delta \kappa_{\alpha\beta} = -\frac{\partial^2 u_3}{\partial x_{\alpha} x_{\beta}}$.

3. The strain field in the plate is approximated as $\varepsilon_{\alpha\beta} = \gamma_{\alpha\beta} + x_3 \cdot \Delta \kappa_{\alpha\beta}$

Where, u_i (*i*=1, 2, 3) are the displacement field for the mid-plane of the nanomembrane in a given coordinate (x_1, x_2, x_3).

3.2.2 Dislocation Mechanics

The misfit dislocation effects on mismatch strain that drives the self-rolling up of strained nanomembranes are examined in detail using atomistic simulations implemented by Lammps and the corresponding desregistry functions of dislocation that describes the relative displacement between two half crystals divided by the heterogenous interface of nanomembranes [39]. In the atomistic simulations, the initial dislocation structure is generated by displacing each atom in simulation box according to the corresponding displacement field of dislocations. Here, we use b, μ and v representing the Burger's vector, shear modulus and Poisson's ratio, respectively. Therefore, the displacement field corresponding to edge dislocations can be written in the following forms in Cartesian coordinates and polar coordinates, respectively [40]:

$$u_x = \frac{b}{2\pi} \left[\tan^{-1} \frac{y}{x} + \frac{xy}{2(1-v)(x^2+y^2)} \right]$$
(3.1)

$$u_{y} = -\frac{b}{2\pi} \left[\frac{1-2v}{4(1-v)} \ln(x^{2} + y^{2}) + \frac{x^{2} - y^{2}}{4(1-v)(x^{2} + y^{2})} \right]$$
(3.2)

$$u_r = \frac{b}{2\pi} \left[-\frac{1-2\nu}{2(1-\nu)} \sin\theta \ln r + \frac{\sin\theta}{4(1-\nu)} + \theta \cos\theta \right]$$
(3.3)

$$u_{\theta} = \frac{b}{2\pi} \left[-\frac{1-2\nu}{2(1-\nu)} \cos\theta \ln r - \frac{\cos\theta}{4(1-\nu)} + \theta \sin\theta \right]$$
(3.4)

The different dislocation configurations can be obtained by changing the center of dislocation displacement field. On the other hand, the desregistry functions of dislocation are calculated through Peierls-Nabarro (PN) model together with an input from the generalized-stacking-fault energy (GSFE) profiles of dislocations, calculated by atomistic simulations [41].

$$K \int_{-\infty}^{\infty} \frac{(du(x)/dx)_{x=x'}dx'}{x-x'} = -\frac{\partial \gamma_{GSFE}(u(x))}{\partial u(x)}$$
(3.5)

where u(x) is the disregistry function describing the relative displacement between two half crystals divided by the slip plane, and *K* is a constant, being $\mu/(4\pi)$ for a screw dislocation and $\mu/[4\pi(1-\nu)]$ for an edge dislocation [42]. The P-N model is deduced by considering the balance between the stress induced by the dislocation (the left side of Eq. (3.5)) and the periodic lattice restoring stress that represents the lattice resistance of a crystal to the distortion associated with the dislocation (the right side of Eq. (3.5)). The disregistry function then could be approximately solved assuming a trial fitting function with undetermined constants α_i and c_i [43]:

$$u(x) = \frac{b}{\pi} \sum_{i=1}^{n} \alpha_i \arctan \frac{x - x_i}{c_i} + \frac{b}{2} , \text{ where } \sum_i^n \alpha_i = 1$$
(3.6)

3.3 Molecular dynamics (MD) simulations

Assuming the validity of classical mechanics and no chemical reactions involved, MD method can simulate the interactions of atoms by constructing an empirical model of interactomic interactions according to the calculated results of quantum mechanics and related experimental data [7]. The whole system would evolve based on the classical equation of motions (Newton's second law)

$$F_i = m_i \frac{\partial^2 r_i}{\partial t^2} \tag{3.7}$$

where m_i represents the mass of the *i*-th atom, the second derivative of the position vector r_i gives the acceleration of the *i*-th atom. F_i is the corresponding force acting on the *i*-th atom, which could be derived from the potential function $V(\{\vec{r}_i\})$ describing the interactions between the atoms, as given:

$$F_i = -\nabla_{\vec{r}_i} V \tag{3.8}$$

The interatomic potential $V(\{\vec{r}_i\})$ is generally expressed as multi-variate series expansion of functional depending on the position vectors \vec{r}_i of atoms and the atom number N involved, which allows the calculation of total energy *E* of the whole system as [8]:

$$V(\{\vec{r}_i\}) = \sum_{i}^{N} V_1(\vec{r}_i) + \sum_{i < j}^{N} V_2(\vec{r}_i, \vec{r}_j) + \sum_{i < j < k}^{N} V_3(\vec{r}_i, \vec{r}_j, \vec{r}_k) + \cdots$$
(3.9)

Where, $V_1(\vec{r_i})$, $V_2(\vec{r_i}, \vec{r_j})$, and $V_3(\vec{r_i}, \vec{r_j}, \vec{r_k})$ represent the one-body term, the two-body or pair potential term and the three-body or multi-body term, respectively, and $V(\{\vec{r_i}\})$ could be further expanded to higher order terms to improve the accuracy of potentials. Here, the summation notions would go over all atoms with the restriction of i < j for the two-body term, i < j < kfor the three-body term and similarly for other higher terms avoiding the double counting of those terms. It is worth noting that due to the quantum nature of interatomic interactions, mathematical approximations are necessary for analytical series expansion of all potentials aiming at improving the computational efficiency, when incorporated into the existing MD codes. Generally, interatomic potentials are developed based on the chosen functional form and varieties with adjustable parameters targeting at different physical properties and some specific material systems. So far, many analytical potentials forms have been developed, such as simple Morse potential [9, 10] and Lennard-Jones (LJ) potential [11] that require less computational power with less physical justifications, and are usually used in the early research, as well as complicated modified embedded-atom method (MEAM) [12] and modified Stilinger-Weber (SW) potentials [13] developed for metals and semiconductors, respectively. Actually, along the advancement of computational technology, interatomic potentials are gradually involving into complicated functional forms with improved precision through fitting more experimental data and quantum simulation results.

Based on the interatomic potential, the MD simulations enable us to obtain full dynamical information of the modelled system that may not be easily observed or detected from experiments. And in this thesis, All the MD simulations are implemented by Software Package Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [14] developed by Sandia National Laboratories, capable of modeling metallic, polymeric, biological systems in a liquid, solid, or gaseous state through applying a wide diversity of interatomic potentials and boundary conditions. Compared with Ab initio calculations, LAMMPS allows dealing with systems containing thousands of atoms, and this is well suited for the study of misfit dislocation effects on the rollup of strained nanomembranes [15, 16]. The interatomic potential used here is modified SW potentials [13, 17] originally developed for InGaN [18, 19]. The general form of total energy of a system for the modified SW potential can be expressed as [17]

$$E = \frac{1}{2} \sum_{i=1}^{N} \sum_{j=i_{1}}^{i_{n}} \left[\varphi_{R,IJ}(r_{ij}) - \varphi_{A,IJ}(r_{ij}) + u_{IJ}(r_{ij}) \times \sum_{\substack{k=i_{1} \\ k \neq j}}^{i_{N}} u_{IK}(r_{ik}) \cdot g_{JIK}(\cos\theta_{jik})^{2} \right] (3.10)$$

Where all adjacent atoms for the center atom *i* are labelled from i_1 to i_n , θ_{jik} indicates the bond angle between atoms *k* and *j* close to the center of atom *i*, and $\varphi_{R,IJ}(r_{ij})$ and $\varphi_{A,IJ}(r_{ij})$ give the pairwise repulsive and attractive functions, respectively, based on the relative position of atoms. $u_{IK}(r_{ik})$ indicates the pair function for three body interactions with $g_{JIK}(cos\theta_{jik})^2$ being the angular penalty function, where the subscripts *i*, *j*, *k* and *I*, *J*, *K*, represent the atom indices and the corresponding species. This potential was generated adopting both experimental data and results from first principle calculations. It can give accurate descriptions of a wide range of structural and material properties of InGaN and allow the detailed investigations of evolution mechanism of misfit dislocations [20] and substitutional diffusion of multi-element alloys [18].

However, there are also several limitations that need to be treated cautiously in MD simulations. First, the material properties related to quantum-mechanics could not be reproduced since the MD method deal with particles on basis of classically Newton's second law instead of Schrodinger equations. Another challenging problem is that the simulations with long time steps (~microsecond) are mathematically ill-conditioning and would induce the numerical instability [21]. Until recently, the simulations up to 1 microsecond were still uncommon and computational intensive especially when large system involved. Additionally, the accuracy of the MD simulations strongly depends on the reliability of the interatomic potentials, and thus special attention should be paid for the potential selected and make sure the potential is transferable and could predict materials properties that are beyond those it was initially approximated and fitted to.

3.4 References

S. Sanchez, Lab-in-a-tube systems as ultra-compact devices, Lab Chip, 15 (2015) 610-613.
 X.L. Li, Self-rolled-up microtube ring resonators: a review of geometrical and resonant properties, Adv Opt Photonics, 3 (2011) 366-387.

[3] I.S. Chun, A. Challa, B. Derickson, K.J. Hsia, X.L. Li, Geometry Effect on the Strain-Induced Self-Rolling of Semiconductor Membranes, Nano Lett, 10 (2010) 3927-3932.

[4] W. Huang, S. Koric, X. Yu, K.J. Hsia, X.L. Li, Precision Structural Engineering of Self-Rolled-up 3D Nanomembranes Guided by Transient Quasi-Static FEM Modeling, Nano Lett, 14 (2014) 6293-6297.

[5] H.S. Lee, M. Cho, B.J. Lee, Size dependence in self-bending mechanism of Ge/Si bilayer nanofilms, J Appl Phys, 109 (2011).

[6] P. Cendula, S. Kiravittaya, I. Monch, J. Schumann, O.G. Schmidt, Directional Roll-up of Nanomembranes Mediated by Wrinkling, Nano Lett, 11 (2011) 236-240.

[7] J.C. Zhang, F. Xu, Y. Hong, Q.G. Xiong, J.M. Pan, A comprehensive review on the molecular dynamics simulation of the novel thermal properties of graphene, RSC Advances, 5 (2015) 89415-89426.

[8] R. LeSar, Introduction to Computational Materials Science: Fundamentals to Applications, Cambridge University Press, 2013.

[9] P.M. Morse, Diatomic molecules according to the wave mechanics. II. Vibrational levels, Phys Rev, 34 (1929) 57-64.

[10] P.M. Morse, Diatomic molecules according to the wave mechanics I: Electronic levels of the hydrogen molecular ion, Phys Rev, 33 (1929) 0932-0947.

[11] J.E. Jones, On the determination of molecular fields - II From the equation of state of a gas, P R Soc Lond a-Conta, 106 (1924) 463-477.

[12] M.I. Baskes, Modified Embedded-Atom Potentials for Cubic Materials and Impurities, Phys Rev B, 46 (1992) 2727-2742.

[13] F.H. Stillinger, T.A. Weber, Computer-Simulation of Local Order in Condensed Phases of Silicon, Phys Rev B, 31 (1985) 5262-5271.

[14] S. Plimpton, Fast Parallel Algorithms for Short-Range Molecular-Dynamics, J Comput Phys, 117 (1995) 1-19.

[15] M.F. Horstemeyer, Integrated Computational Materials Engineering (ICME) for Metals: Using Multiscale Modeling to Invigorate Engineering Design with Science, Wiley, 2012.

[16] W. Hayes, A.M. Stoneham, Defects and Defect Processes in Nonmetallic Solids, Dover Publications, 2012.

[17] X. Zhou, M.E. Foster, R. Jones, P. Yang, H. Fan, F. Doty, A Modified Stillinger-Weber Potential for TIBr, and Its Polymorphic Extension, Journal of Materials Science Research, 4 (2015) 15.

[18] X.W. Zhou, R.E. Jones, J. Gruber, Molecular dynamics simulations of substitutional diffusion, Comp Mater Sci, 128 (2017) 331-336.

[19] X.W. Zhou, R.E. Jones, K. Chu, Polymorphic improvement of Stillinger-Weber potential for InGaN, J Appl Phys, 122 (2017) 235703.

[20] J. Gruber, X.W. Zhou, R.E. Jones, S.R. Lee, G.J. Tucker, Molecular dynamics studies of defect formation during heteroepitaxial growth of InGaN alloys on (0001) GaN surfaces, J Appl Phys, 121 (2017).

[21] S. Li, X.L. Gao, Handbook of Micromechanics and Nanomechanics, Pan Stanford Publishing, 2016.

[22] T. Huang, Z.Q. Liu, G.S. Huang, R. Liu, Y.F. Mei, Grating-structured metallic microsprings, Nanoscale, 6 (2014) 9428-9435.

[23] D. Grimm, C.C.B. Bufon, C. Deneke, P. Atkinson, D.J. Thurmer, F. Schaffel, S. Gorantla, A. Bachmatiuk, O.G. Schmidt, Rolled-up nanomembranes as compact 3D architectures for field effect transistors and fluidic sensing applications, Nano Lett, 13 (2013) 213-218.

[24] J. Zang, F. Liu, Modified Timoshenko formula for bending of ultrathin strained bilayer films, Appl Phys Lett, 92 (2008).

[25] J.N. Reddy, D.K. Gartling, The Finite Element Method in Heat Transfer and Fluid Dynamics, Third Edition, Taylor & Francis, 2010.

[26] P. SESHU, TEXTBOOK OF FINITE ELEMENT ANALYSIS, PHI Learning, 2003.

[27] E.J. Barbero, Finite Element Analysis of Composite Materials using AbaqusTM, Taylor & Francis, 2013.

[28] G. Puri, Python Scripts for Abaqus: Learn by Example, Kan sasana Printer, 2011.

[29] E.J. Barbero, Finite Element Analysis of Composite Materials, Taylor & Francis, 2007.

[30] A. Khennane, Introduction to Finite Element Analysis Using MATLAB® and Abaqus, Taylor & Francis, 2013.

[31] K. Hibbitt, Sorensen, ABAQUS/Standard User's Manual, Hibbitt, Karlsson & Sorensen, 2001.

[32] ABAQUS., K. Hibbitt, Sorensen, ABAQUS Explicit: User's Manual : Volume 2 : Version 5.8, Hibbitt, Karlsson and Sorensen, 1998.

[33] S.S. Rao, The Finite Element Method in Engineering, Elsevier Science, 2017.

[34] C. Teodosiu, Large Plastic Deformation of Crystalline Aggregates, Springer Vienna, 2014.[35] A.F. Bower, Applied Mechanics of Solids, CRC Press, 2009.

[36] D. Hull, D.J. Bacon, Introduction to Dislocations, Elsevier Science, 2011.

[37] S. Vidoli, Discrete approximations of the Foppl-Von Karman shell model: From coarse to more refined models, Int J Solids Struct, 50 (2013) 1241-1252.

[38] O.A. Bauchau, J.I. Craig, Kirchhoff plate theory, in: O.A. Bauchau, J.I. Craig (Eds.) Structural Analysis, Springer Netherlands, Dordrecht, 2009, pp. 819-914.

[39] B. Joos, Q. Ren, M.S. Duesbery, Peierls-Nabarro Model of Dislocations in Silicon with Generalized Stacking-Fault Restoring Forces, Phys Rev B, 50 (1994) 5890-5898.

[40] P.M. Anderson, J.P. Hirth, J. Lothe, Theory of Dislocations, Cambridge University Press, 2017.

[41] V. Bulatov, W. Cai, Computer Simulations of Dislocations, OUP Oxford, 2006.

[42] C. Sansour, S. Skatulla, Generalized Continua and Dislocation Theory: Theoretical Concepts, Computational Methods and Experimental Verification, Springer Vienna, 2012.

[43] V.V. Bulatov, E. Kaxiras, Semidiscrete variational Peierls framework for dislocation core properties, Phys Rev Lett, 78 (1997) 4221-4224.

Chapter 4: Quantitative Analysis and Predictive Engineering of Self-rolling of Nanomembranes Under Anisotropic Mismatch Strain

This thesis starts with the investigation of the dependence of self-rolling behaviors of nanomembranes on the anisotropic mismatch strain. It mainly focusses on the precise evaluation and manipulation of rollup curvature and direction of strained nanomembranes through continuum theory and FE analysis. Subsequently, based on the analytical and simulated results, several microfabrication strategies have been proposed to predictively manipulate the rolling direction of strained nanomembranes and optimize the roll-up yield, which provides important guidelines for design and prediction of novel 3D nanomembrane geometries.

• This chapter has been published in *Nanotechnology*, appeared as:

Quantitative analysis and predictive engineering of self-rolling of nanomembranes under anisotropic mismatch strain.

Cheng Chen, Pengfei Song, Fanchao Meng, Xiao Li, Xinyu Liu, Jun Song* DOI: https://doi.org/10.1088/1361-6528/aa94aa

4.1 Abstract

The present work presents a quantitative modeling framework for investigating the selfrolling of nanomembranes under different lattice mismatch strain anisotropy. The effect of transverse mismatch strain on the roll-up direction and curvature has been systematically studied employing both analytical modeling and numerical simulations. The bidirectional nature of the self-rolling of nanomembranes and the critical role of transverse strain in affecting the rolling behaviors have been demonstrated. Two fabrication strategies, i.e., third-layer deposition and corner geometry engineering, have been proposed to predictively manipulate the bidirectional rolling competition of strained nanomembranes, so as to achieve controlled, unidirectional roll-up. In particular for the strategy of corner engineering, microfabrication experiments have been performed to showcase its practical application and effectiveness. Our study offers new mechanistic knowledge towards understanding and predictive engineering of self-rolling of nanomembranes with improved roll-up yield.

4.2 Introduction

The tubular and helical-shaped nanomembrane structures have found wide applications in biomedical devices [1-4], optoelectronics [5-7] and nano/micro-electromechanical components [8-10], thanks to their great versatility and tunable functionality [11-13]. It has been demonstrated that the precisely controlled rolled-up helical structures with pre-designed helical angle, pitch, chirality as well as diameter allow the tuning of their optical and mechanical properties and promise important applications in the next generation of micro-/nano-fluidic sensors and actuators [12-15]. A widely used strategy for creating such nanomembrane structures is based on strain engineering, which utilizes built-in mismatched strains to drive initially flat, one- or multilayer nanomembranes to self-roll into 3D tubular or helical structures [16-19]. There have been extensive research efforts devoted to developing controllable fabrication processes so as to achieve precise geometric designs of the rolled-up structures [20-25]. However, the rolling-up mechanics of these structures is still not fully understood, even for the roll-up of nanomembranes of the simple rectangular configuration. It has been revealed that as rectangular nanomembranes are released from their substrate through etching off the underneath sacrificial layer, the isotropic releasing of in-plane strain would induce equal tendency of the nanomembranes to roll up from all four edges and thus lead to competition among the two potential roll-up directions [20, 25]. Such competition results in great uncertainties in the determination of not only the dominant roll-up direction, but also the final curvature of the rolled-up structure.

Many theoretical studies [26-28] have been conducted, attempting to model the roll-up process and predict the resultant curvature. Yet despite those efforts, there is still considerable discrepancy between the model predicted and experimentally observed roll-up curvatures. Various possibilities have been proposed to explain the discrepancy, including nonlinear strain effect [29], surface tension and reconstruction [30], thickness deviation and extra strain [31,
32]. However, such explanation is mostly qualitative and inconsistency exists [18, 22, 33], and there lacks a quantitative method to analyze the discrepancy and to evaluate contributions from different effects. Meanwhile, it remains a great challenge to obtain desirable unidirectional rollup configurations [22, 25, 34]. It has been revealed that the roll-up can occur predominantly along either the longitude or transverse direction, or simultaneously along both. This however leads to randomness in the resultant roll-up structure, producing different roll-up geometries including, for instance, "tick or check mark-like" structures [35] induced by the diagonal rolling and all edges rolling, which greatly depends on the length and width of the nanomembrane and the history of rolling process as well as the diameter of the roll-up tubes [25, 35]. Given that the strong dependence of device performance fabricated by the tubular or helical-shaped nanomembrane on the roll-up geometry, including photons interaction and mechanical properties [12-15], this randomness disrupts the fabrication of nano-devices with precisely controllable dimensions and position. Several methods have been proposed, attempting to control the resultant roll-up direction, including the introduction of wrinkles geometry [20], patterning (e.g., grating structures [23] and hollow circles [25]), angle deposition[16], and lithographically deep etching [36, 37]. However, because of the complex geometrical features required for the films, those methods often suffer from issues of local damages and/or ruptures induced by the multiple etching processes and photolithography required [38, 39], and deposition inaccuracy [20].

In the present study, starting from Von-Karman plate theory [40, 41] and Rayleigh-Ritz model [40, 42, 43], we obtained a series of formulas for analytical modeling of the bi-directional roll-up of nanomembranes, accounting for anisotropic lattice mismatch. The effect of transverse mismatch strain on the roll-up direction and curvature has been systematically studied employing both analytical modeling and numerical simulations. It is demonstrated that the transverse strain can be a critical factor contributing to the discrepancy between previous model predictions and experimental measurements. Motivated by our modeling and simulation results, two fabrication strategies, i.e., third-layer deposition and corner engineering, have been proposed to predictively manipulate the roll-up process of strained nanomembranes. In particular for the strategy of corner engineering, microfabrication experiments have been performed to showcase its practical application and effectiveness. Our study offers new mechanistic knowledge towards understanding and predictive engineering of self-rolling of nanomembranes with improved roll-up yield.

4.3. Methodology

4.3.1 Analytical formulation

In our study, we consider a rectangular nanomembrane of in-plane dimensions L in length (x_1 direction) and W in width (x_2 direction), comprising two layers bonded together as illustrated in Fig. 4.1a, a typical precursor geometry used in experiments to produce roll-up structures. The top and bottom layers are respectively of thicknesses (h_t , h_b), Poisson ratio (v_t , v_b), and Young's moduli (E_t , E_b). The mid-plane of the nanomembrane is thus at height ($h_t + h_b$)/2 from the base of the bottom film. Assuming the presence of lattice mismatch between the two layers, with mismatch strain ε_0 and $\eta \varepsilon_0$ along the x_1 and x_2 directions respectively, where η represents the anisotropy of lattice mismatch, the nanomembrane will undergo deformation. An approximated displacement field, i.e., u_i (*i*=1, 2, 3) of the mid-plane of the nanomembrane can be obtained based on the Ritz method [40, 42, 43] as:

$$u_1 = C_1 x_1 + C_2 x_1^3 + C_3 x_1 x_2^3 \tag{4.1}$$

$$u_2 = C_4 x_2 + C_5 x_2^3 + C_6 x_2 x_1^2 \tag{4.2}$$

$$u_3 = \frac{1}{2}(ax_1^2 + bx_2^2) \tag{4.3}$$

where C_i (*i*=1-6), *a* and *b* are eight parameters to be determined. From the displacement fields, the rolling curvatures can be further calculated as:

$$\kappa_{11} = \frac{\partial^2 u_3}{\partial x_1^2} \text{ and } \kappa_{22} = \frac{\partial^2 u_3}{\partial x_2^2}$$
(4.4)

Below we refer to roll-up along length and width directions as longitude and transverse directions respectively, and in accordance κ_{11} and κ_{22} as longitude and transverse curvatures. On the other hand, in the strained system, the elastic strain fields in the two layers are determined through displacement field using the Von-Karman formula [41]:

$$\varepsilon_{\alpha\beta} = \frac{1}{2} \left(\frac{\partial u_{\alpha}}{\partial x_{\beta}} + \frac{\partial u_{\beta}}{\partial x_{\alpha}} + \frac{\partial u_{3}}{\partial x_{\beta}} \cdot \frac{\partial u_{3}}{\partial x_{\alpha}} \right) - x_{3} \frac{\partial^{2} u_{3}}{\partial x_{\alpha} \partial x_{\beta}}$$
(4.5)

with α , $\beta = 1$ or 2. By incorporating contributions from inelastic mismatch strains, i.e., ε_0 and $\eta \varepsilon_0$ respectively along the x_1 and x_2 directions, we can then obtain total strain field. With the rectangular nanomembrane assumed to be in a state of plane-stress, the stress components of substrate and film($\sigma_{ij}^b, \sigma_{ij}^t$) can be determined, and the total potential energy of the system can be evaluated as

$$U = \iiint_V \left(\frac{1}{2}\sigma^b_{ij}\varepsilon^b_{ij} + \frac{1}{2}\sigma^t_{ij}\varepsilon^b_{ij}\right)dV$$
(4.6)

The coefficients and curvatures can then be determined by minimizing the potential energy U. For simplicity in derivation and analysis, in the following, the condition of W = L is assumed.

A closed-form relationship between longitudinal and transverse curvatures can be obtained for the special case of two layers of the same Poisson ratio, i.e., $v_b = v_t = v$, and $\eta =$ 1, as shown below

$$(\kappa_{11} - \kappa_{22}) \left(\kappa_{11} \kappa_{22} - \frac{3}{2} \frac{(v+6) \left(h_t^4 E_t^2 + 4h_t \left(h_t^2 + \frac{3}{2} h_b h_t + h_b^4 \right) E_b E_t h_b + h_b^4 E_b^2 \right)}{L^4 (E_b h_b + E_t h_t)^2 (v+1)} \right) = 0$$
(4.7)

Eq. 4.7 indicates that multiple equilibrium states for the bidirectional roll-up exist, as illustrated in Figs. 4.1b-c, in the two categories of (I) $\kappa_{11} = \kappa_{22}$, and (II) the two curvatures being related as



$$\kappa_{11}\kappa_{22} = \frac{3}{2} \frac{(\nu+6)\left(h_t^4 E_t^2 + 4h_t\left(h_t^2 + \frac{3}{2}h_b h_t + h_b^4\right)E_b E_t h_b + h_b^4 E_b^2\right)}{L^4 (E_b h_b + E_t h_t)^2 (\nu+1)}$$
(4.8)

Figure 4.1. (a) Schematic illustration of a strained rectangular bilayer nanomembrane of length *L* and width *W*, with mismatch strain ε_0 and $\eta \varepsilon_0$ along the x_1 (longitude) and x_2 (transverse) directions respectively. The top and bottom layers are respectively of thicknesses (h_t , h_b), Poisson ratio (v_t , v_b), and Young's moduli (E_t , E_b). Multiple equilibrium states exist for the bidirectional roll-up of the nanomembrane, in two categories, i.e., (b) of equal curvatures along longitude and transverse directions, $\kappa_{11} = \kappa_{22}$, and (c) with the two curvatures related via Eq. 4. 8.

The explicit expressions of curvatures can be obtained if we further assume that the two layers of are the same thickness, i.e., $h_t = h_b = h$, yielding:

Category (I):

$$\kappa_{11} = \kappa_{22} = \frac{2^{4/3} \left(\frac{1}{4} \left(h(\nu+6) \left(9L^2 \varepsilon_0 + A\right)(\nu-1)^2\right)^{2/3} + \left(\nu^2 + 5\nu - 6\right)h^2 2^{1/3}\right)}{(h(\nu+6) \left(9L^2 \varepsilon_0 + A\right)(\nu-1)^2\right)^{1/3} L^2(\nu-1)},\tag{4.9}$$

where
$$A = \sqrt{\frac{81L^4(\nu-1)\varepsilon_0^2 - 128h^4(\nu+6)}{\nu-1}}$$
.

Category (II):

$$(\kappa_{11}, \kappa_{22}) = (\kappa^+, \kappa^-) \text{ or } (\kappa_{11}, \kappa_{22}) = (\kappa^-, \kappa^+)$$
 (4.10)

with

$$\kappa^{+} = \frac{\sqrt{3}}{8} \frac{\sqrt{\frac{3L^{4}(\nu+1)^{3}\varepsilon_{0}^{2}-128h^{4}(\nu+6)}{\nu+1}} + 3\varepsilon_{0}(\nu+1)L^{2}}{L^{2}h}$$
(4.11)

$$\kappa^{-} = \frac{\sqrt{3}}{8} \frac{\sqrt{\frac{3L^{4}(\nu+1)^{3}\varepsilon_{0}^{2}-128h^{4}(\nu+6)}{\nu+1}} - 3\varepsilon_{0}(\nu+1)L^{2}}{L^{2}h}$$
(4.12)

From Eqs. 4.11-12, we can see that in the particular case of very thin film, i.e., $h \ll L$ (and W), the curvature $\kappa^- \rightarrow 0$, being negligible compared to κ^+ , and consequently the roll-up effectively becomes unidirectional.

The three equilibrium states (i.e., one in Category I and two in Category II) presented in Fig. 4.1, are generally expected during the self-rolling of nanomembranes, namely, (*A*) two comparable curvatures, (*B*) dominant longitudinal roll-up with minimal transverse curvature, (*C*) dominant transverse roll-up with minimal longitudinal curvature. It is noteworthy that in the above derivations, the condition of $\eta = 1$ was assumed to get the analytical forms of curvatures (in Eqs. 4.9-12) and explain the possible three equilibrium states, because the explicit expressions of curvatures for multiple equilibrium states cannot be obtained for all η values. For material systems with other η values (e.g., the cases of $\eta = 0.25$, 0.5 and 0.75 shown in Fig. 2), the numerical approach needs to be used (to solve Eqs. 4.1-6). This is further illustrated in Fig. 4.2, for the representative bilayer GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom), a material system widely used to fabricate self-assembly semiconductor nanostructures [44-46]. The mechanical properties [17] are taken as $(E_t, E_b) = (85.6, 75.1)$ GPa, $v_t = v_b = 0.3$. The mismatch strain along the length direction (i.e., longitude direction) is set as 1.43×10^{-2} , while that along the width direction (i.e., transverse direction) varies, being $1.43\eta \times 10^{-2}$ with $\eta = 0.25$, 0.5, 0.75 and 1. In our calculations, *L* and *W* were set to be much larger than the roll-up diameter so that the results are independent of the in-plane dimensions. The thickness *h* is well controlled to achieve different strain gradients. The curvature pairs corresponding to the three equilibrium states are denoted as $(\kappa_{11}^A, \kappa_{22}^A), (\kappa_{11}^B, \kappa_{22}^B)$ and $(\kappa_{11}^C, \kappa_{22}^C)$ in Fig. 4.2. Several key observations can be drawn from Fig. 4.2. We note that there is a stability transition, from the multiple equilibrium states or less, state *A* becomes unstable while states *B* and *C* prevail. Since the typical nanomembrane thickness for roll-up nanostructures is < 1 µm, we focus our discussion on states *B* and *C* in the follows.



Figure 4.2. Stability diagrams illustrating multiple equilibrium states for κ_{11} and κ_{22} with different level of lattice mismatch anisotropy (η): (a) $\eta = 0.25$, (b) $\eta = 0.5$, (c) $\eta = 0.75$, and (d) $\eta = 1$. With fixed *W* and *L* (in this particular example, we set $L = W = 100 \,\mu\text{m}$), the *h* is well controlled to achieve different strain gradients and thus varied equilibrium states. The resulting curvature pairs corresponding to the three equilibrium states (see Section 4.2.1 in the text) are denoted as ($\kappa_{11}^A, \kappa_{22}^A$), ($\kappa_{11}^B, \kappa_{22}^B$) and ($\kappa_{11}^C, \kappa_{22}^C$).

We can further note that under the condition of $h < 1 \mu m$, one roll-up curvature (corresponding to state *B* or *C*) is approximately zero. Assuming $\kappa_{11} = 0$ or $\kappa_{22} = 0$, we can obtain explicit solutions of curvatures in more general conditions as the following with $\rho = \frac{h_t}{h_h}$ and $E_r = \frac{E_t}{E_h}$

$$\kappa_{22} = 0, \ \kappa_{11} = \frac{6 \cdot \varepsilon_0 \cdot E_r \cdot \rho \cdot (\rho + 1)(1 + \eta \nu)}{h_b \left(\rho^4 + 4\rho \cdot \left(\rho^2 + \frac{3}{2}\rho + 1\right)E_r + E_r^2\right)}$$
(4.13)

$$\kappa_{11} = 0, \ \kappa_{22} = \frac{6 \cdot \varepsilon_0 \cdot E_r \cdot \rho \cdot (\rho + 1)(\nu + \eta)}{h_b \left(\rho^4 + 4\rho \cdot \left(\rho^2 + \frac{3}{2}\rho + 1\right)E_r + E_r^2\right)}$$
(4.14)

It is also worth to note that in practice, once one roll-up direction prevails, it will effectively render the curvature of the other direction zero. Thus, the above can be regarded as being directly consequent of states *B* and *C*. Eqs 4.13 and 4.14 are essential equations to understand the competition between roll-up along longitudinal and transverse directions. They clearly elucidate the bi-directional nature of the nanomembrane roll-up and indicate the necessity to consider the transverse effect when assessing the final roll-up geometry. It is also clear from these equations that there is a strong dependence on the parameter η . Note that in the particular case of $\eta = 0$, Eq. 4.13 simplifies to the Timoshenko formula [47], which was employed by many previous studies to predict the roll-up curvature. As elaborated below (Section 3), we demonstrate that the Timoshenko formula is often insufficient and can result in sizable discrepancy between experimental measurements and theoretical predictions, and that our study offers a new analytical method that yields much closer estimations to experimental results.

4.3.2 Roll-up geometries from numerical simulations

To validate the analytical predictions of the roll-up curvatures, we perform finiteelement (FE) simulations, using the ABAQUS Unified FEA software [48], to analyze the rollup process numerically. The strained bilayer structure was built based on the module of composite layups, and the 4-node doubly curved thin/thick shell element with reduced integration (S4R) was used. The variation of mismatch strain was achieved by assigning different thermal expansion coefficients to materials and varying temperature. Quasi-static method was employed to examine the roll-up process under controlled release, i.e., the strained bilayers are partitioned into strips and fixed initially, and then released gradually. The roll-up direction can be further controlled by clamping one side initially with a delayed release. The controlled roll-up simulation mimics the situation where etching occurs only or preferentially along one direction. It is worth noting that for those controlled roll-up simulations, once a considerable degree of roll-up occurs along the intended direction (long or short), the roll-up will continue even after the artificial clamping of the other side is released. This means the roll-up of strained nanomembrane is a strongly history-dependent process, well agreed with the previous experimental observations [25]. Meanwhile ABAQUS explicit/dynamic package has been used to conduct dynamic simulations of cases of simultaneous release (i.e., no preconstraint, corresponding to the situation of isotropic etching) to study the competition between different roll-up directions.

4.4 **Results and Discussion**

4.4.1 FE simulated and analytically predicted roll-up curvatures

Comprehensive FE simulations have been performed to examine the roll-up of nanomembranes of different combinations of material parameters and control conditions. We first examine the longitude roll-up of nanomembrane (illustrated in Fig. 4.3a), the commonly preferred situation in experiments [21, 49]. Fig. 4.3 presents some representative plots illustrating the FE simulated and analytically predicted (cf. Eqs. 4.13-14) evolution of the roll-up diameter *D* (related to curvature via $D = 2/\kappa$), as the mismatch strain ε_0 , Young's modulus E_t and film thickness h_t vary, for different degrees of anisotropy (i.e., $\eta = 0, 0.25, 0.5$ and 1) of lattice mismatch. The reference values of strain, modulus and film thickness used in our calculations are taken from previous studies [17, 18, 32] for the representative system of GaAs/In_xGa_{1-x}As, on top of which additional variation of those parameters that possibly originates from, e.g., changes in composition (i.e., In_xGa_{1-x}As of different *x*) or thickness of the

bottom layer. Shown in Fig. 4.3, the predicted roll-up diameter D decreases as the mismatch strain increases while increases as the modulus and film thickness increase. Meanwhile, we note that larger values of η leads to smaller roll-up curvatures, indicating that the transverse mismatch strain negatively impacts the longitude roll-up. The FE simulation data are in excellent agreement with the analytical results, a good indication of the accuracy and predictive capacity of our analytical formulas.



Figure 4.3. (a) A schematic illustration of the FE simulated roll-up configuration of a bilayer nanomembrane. With the longitude roll-up assumed and the material parameters set with respect to the representative system of GaAs/In_xGa_{1-x}As, the FE simulated and analytically predicted evolutions of the resultant roll-up diameter *D* as (b) the longitudinal mismatch strain ε_0 , (c) Yong's modulus of top layer E_t , and (d) the thickness of top layer h_t vary, under the influence of different level of lattice mismatch anisotropy (η).

4.4.2 Implications to practical applications

4.4.2.1 Accurate assessment of the roll-up curvature

Most previous theoretical evaluations [17, 18, 22, 32, 33, 49, 50] of the roll-up curvature are based on the classical Timoshenko formula where the transverse strain was not considered. As unidirectional mismatch is not the case for most material systems, the usage of the Timoshenko formula would unavoidably result in discrepancy between experimental measurements and theoretical prediction, as confirmed by many prior studies [17, 32, 51]. Fig. 4.4 compares the experimentally measured and theoretically predicted values of roll-up diameter D, for two nanomembrane systems, the $GaAs^{(top)}/InAs^{(bottom)}$ [29, 32] and the $SiN_x^{(top)}/SiN_x^{(bottom)}$ [21] systems. As seen in Fig. 4.4, for both cases the Timoshenko formula considerably overestimates the curvature. In contrast, good match between experimental results and our model predictions (from Eq. 4.13) can be achieved with the anisotropy of lattice mismatch η between 1 and 1.25. The non-zero values of η clearly indicates the necessity of accounting for the effect of transverse strain on the roll-up. This mismatch strain anisotropy has also been experimentally confirmed [52-55]. As previously mentioned, in the previous studies the discrepancy between experimental measurements and theoretical prediction was often conveniently attributed to effects such as oxidation formation [32] additional strain [17, 18] or surface tension [30], while the influence of transverse strain was not well accounted for. Our results identify transverse strain as another important factor that contributes to the discrepancy. Furthermore, the results also indicate the interesting possibility of using the transverse strain as a generic metric (that can incorporate other effects) to quantitatively analyze the discrepancy.



Figure 4.4. Comparison between theoretically predicted (lines) and experimentally measured (open symbols) roll-up diameters for the (a) $GaAs^{(top)}/InAs^{(bottom)}$ nanomembrane [32] and (b) $SiN_x^{(top)}/SiN_x^{(bottom)}$ nanomembrane [21], as the thickness of the top layer varies. The parameter η denotes the anisotropy of lattice mismatch. The predictions from Timoshenko formula (dotted lines) and the previous study by Grunmann [29] (pink dash-dot-dot line in (a)) are also included for comparison.

4.4.2.2 Design ideas towards predictively control of the roll-up direction

As previously discussed, the roll-up of a rectangular nanomembrane is bidirectional in nature. Therefore, it is of great importance to be able to control the roll-up direction aiming at realizing of desirable structure and geometry. Motivated by our modeling results above, below we propose two design strategies, based on third-layer deposition and corner geometry engineering, to accurately manipulate the roll-up direction. Note that in the content below we designate the longitude roll-up to be the desired outcome.

4.4.2.2.1 Third-layer deposition

Since the root cause of the bi-directional competition is the transverse mismatch strain, an apparent method to achieve the desired longitude roll-up is to moderate or even eliminate the transverse strain. Ideally this can be realized by depositing an extra third-layer, which can be designed to have an in-plane mismatch strain of the opposite sign as the transverse strain existing between the bottom and top layers (of the original bilayer system), thereby producing a bending moment M_1 that counteracts the transverse effect, as illustrated in Fig. 4.5.



Figure 4.5. Schematic illustration of predictive control of the roll-up direction through the deposition of an additional third layer. (a) Prestrained bilayer exhibiting mixed roll-up configurations consequent of the bi-directional competition. (b) Rolling competition suppressed by the extra third layer.

The magnitude of M_1 can be tuned by adjusting the thickness of the third layer. The role of third-layer deposition on roll-up can be incorporated and analytically treated by extending the bilayer model. Denoting the Young's modulus, thicknesses, and Poisson's ratio of the third layer as E_3 , h_3 , and v_3 , and for simplicity but without loss of generality, assuming that the third-layer results in an isotropic mismatch strain ε_1 and that the three layers exhibit the same Poisson's ratio and Young's modulus, $v_b = v_t = v_3 = v$ and $E_b = E_t = E_3 = E$, the curvatures corresponding to unidirectional roll-up along longitude and transverse directions can be obtained following a similar process carried out for the bilayer system (c.f. Section 2.1)

Longitude roll-up:

$$\kappa_{22} = 0, \kappa_{11} = \frac{\left((6\eta\epsilon_1\nu - 6\epsilon_0\nu - 6\epsilon_0 + 6\epsilon_1)h_t - 6\epsilon_1h_bh_t(\eta\nu + 1)\right) - 6\epsilon_0h_3h_s(\nu + 1)}{(h_3 + h_t + h_b)^3}$$
(4.15)

Transverse roll-up:

$$\kappa_{11} = 0, \, \kappa_{22} = \frac{\left((6(\eta+\nu)\varepsilon_1 - (6\nu+6)\varepsilon_0)h_t - 6\varepsilon_1h_bh_t(\eta+\nu)\right) - 6\varepsilon_0h_3h_b(\nu+1)}{(h_3 + h_t + h_b)^3} \tag{4.16}$$

Comparing the above two equations to Eqs. 4.13 and 14 in Section 4. 2.1, we see a new degree of freedom, the third-layer thickness h_3 , which could be adjusted experimentally to tune the competition between longitude and transverse directions, and their corresponding curvatures, κ_{11} and κ_{22} . Specifically, the critical third layer thickness that eliminates the transverse effect, i.e., at which the curvature κ_{22} for transverse roll-up becomes zero, is

$$h_3^c = -\frac{\varepsilon_0 h_t h_b(\nu + \eta)}{(h_t + h_b)(1 + \nu)\varepsilon_0 - \varepsilon_1(\nu + \eta)h_t}$$
(4.17)

At $h_3 = h_3^c$, the rolling is only viable along the longitude direction, and the corresponding curvature is

$$\kappa_{11}^{c} = \frac{6(\nu^{2}-1)(((\eta+\nu)\varepsilon_{1}-\varepsilon_{0}(\nu+1))h_{t}-\varepsilon_{0}h_{b}(1+\nu))^{2}(\eta-1)h_{t}h_{b}(h_{t}+h_{b})\varepsilon_{1}\varepsilon_{0}}{(((\eta+\nu)\varepsilon_{1}-\varepsilon_{0}(\nu+1))h_{t}^{2}+2h_{b}((\eta+\nu)\varepsilon_{1}-\varepsilon_{0}(\nu+1))h_{t}-\varepsilon_{0}h_{b}^{2}(1+\nu))^{3}}$$
(4.18)

To assess the feasibility of third-layer deposition, below we performed an analytical demonstration for a model three-layer nanomembrane, where the original top and bottom layers are of thicknesses $(h_b, h_t) = (50\text{\AA}, 100\text{\AA})$, Young's modulus of 80 GPa and Possion's ratio of 0.3, with a mismatch $\varepsilon_0 = 1.43 \times 10^{-2}$ and different levels of mismatch anisotropy $\eta = 0.25$, 0.5 or 0.75. Fig. 4.6 shows how the longitude (κ_{11}) and transverse (κ_{22}) curvatures evolve as the third-layer thickness changes, at different levels of mismatch anisotropy η (cf. Fig. 4.6a)

and third-layer induced isotropic mismatch strain ε_1 (cf. Fig. 4.6b). We can see that a thirdlayer of just a few nanometers in thickness can have profound impact on the roll-up curvature, demonstrating the capability of third-layer deposition in tuning the longitude-transverse competition and resultant roll-up curvature.



Figure 4.6. The evolution of bidirectional roll-up curvatures κ_{11} and κ_{22} predicted from Eqs 4.15-16 as the function of the third layer thickness h_3 , under different levels of (a) lattice mismatch anisotropy η (with $\varepsilon_1 = -\varepsilon_0$) and (b) isotropic mismatch strain ε_1 (with $\eta = 0.5$) for a model three-layer nanomembrane (see details in text).

It is, however, important to recognize the third-layer deposition simultaneously reduces curvatures for both longitude and transverse roll-ups, as illustrated in Fig. 4.6, which can be undesirable in the design. Nonetheless, this limitation can be prevented if simultaneous adjustment of thicknesses of all three layers is possible, in which case we can introduce a prefactor S_d , being $S_d = \kappa_{11}^c / \kappa_{11}$ (with κ_{11} and κ_{11}^c defined in Eqs. (4.16) and (4.18) respectively), and scale down the thicknesses of the three layers as $(h_b, h_t, h_3^c) \rightarrow S_d \cdot (h_b, h_t, h_3^c)$. This way we can avoid curvature reduction and maintain the curvature expected from the original bilayer nanomembrane. In fact, the above scaling down approach can also be extended to enable us to achieve an arbitrary curvature (κ_{11}^A) by changing the scale factor (i.e., simply set $S_d = \kappa_{11}^c / \kappa_{11}^A$). The third layer deposition method is expected to be applied for the material systems being mismatch strain anisotropy (i.e., GaN/ZnO [56], either in the original bilayer system or the deposited third-layer). Such material systems are usually synthesized by the expensive and sophisticated equipment such as the molecular beam epitaxy (MBE) and/or metalorganic chemical vapour deposition (MOCVD), which are not accessible to the authors currently, preventing us from conducting direct experimental validation.

4.4.2.2.2 Corner geometry engineering

Under the condition of isotropic etching, interface regions of large local curvatures (e.g., corners) are under more "*attack*" [20, 57, 58], thus representing the critical locations affecting the roll-up process, particularly at the initial stage. Therefore, one may manipulate the process through modifying the corner geometry. Here we propose a design strategy based on corner geometry engineering, where corner attachments are introduced to the nanomembranes to tilt the competition between longitude and transverse directions to realize the unidirectional (longitude) roll-up. The concept of corner engineering is illustrated for two sample (i.e., rectangular and U-shape) nanomembrane geometries in **Fig. 7**, showing their configurations without (cf. Figs 4.7a and c) and with (cf. Figs 4.7b and d) corner attachments. Here the U-shape configuration is included because it is one of the most common design layouts used for many applications, such as microtube lasers [45] nanoparticles detection [59] and liquid sensing [60] due to its capability of lifting-up the middle part of the microtube from the substrate after rolling-up [61]. It is worthy to note that the U-shape configuration often involves the one-end clamping constrain, corresponding to the situation of a cover layer existing to prevent etching from one end [45, 62], which is also reflected in Fig. 4.7.

FE simulations were then conducted on those nanomembranes (with and without corner attachments) to examine their rolling behaviors. The bilayer $SiN_{x'}^{(top)}/SiO_{x}^{(bottom)}$ is selected as the representative material system, which is also the one used in our experimental validation

(see below). The thicknesses of the SiN_x and SiO_x layers, (h_b, h_t) , are set as (150Å, 200Å), the elastic properties, i.e., Young's moduli and Poisson ratio (E_b, E_t, v) are taken as (75GPa, 180 GPa, 0.25), and an isotropic mismatch strain of 5.4×10^{-2} is assumed [49]. The dimensions of the initial flat nanomembranes are given in Figs 4.7a and 7c. For simplicity, a rectangular corner attachment of the same properties as the bilayer nanomembrane is considered. As seen in Figs 4. 7a and 7c, for the nanomembranes free of corner attachments, the rolling initiates largely from the transversal direction (See videos in Electronic Supplementary Information), as observed previously [20, 25]. It is attributed to the nonuniform spatial distribution of curvatures and doubly curved regions at the corner of strained nanomembranes [24, 58]. In contrast, as demonstrated in Figs 4.7b and d, the desirable longitude roll-up can be achieved when aided by corner engineering. A more comprehensive examination reveals that such control of roll-up direction requires the corner attachment to satisfy certain geometrical constrains. This requirement is demonstrated in Fig. 4.8a and b for the particular case of rectangular corner attachments, where the length-width regimes corresponding to different rolling scenarios are identified. We see that (for a specific width) the length/width ratio of the corner attachment needs to exceed certain threshold value in order to achieve unidirectional longitude roll-up.



Figure 4.7. Schematic illustration of the role of corner attachment in achieving controlled unidirectional roll-up along the longitude direction, for nanomembranes. The figures in left and right columns present the initial and resultant FE-simulated roll-up configurations respectively, showing the involvement of transverse rolling mode for (a) rectangular and (c) U-shape nanomembranes without corner attachments, and the realization of desirable longitude roll-up for (b) rectangular and (d) U-shape nanomembranes (See videos in Electronic Supplementary Information). The red segments in (c) and (d) indicate the clamped regions in U-shape nanomembranes.



Figure 4.8. Diagram of preferred rolling direction as the length and width of the rectangular corner attachment vary, the regimes where long-edge (transverse) and short-edge (longitude) rolling occur are colored white and grey respectively, for (a) rectangular and (b) U-shape nanomembranes. The star symbol indicates the length/width of the corner attachment used in the microfabrication experiments of the rolled-up microtube in (c-f). (c) An Al sacrificial layer is deposited on a silicon wafer, on top of which (d) a SiN_x^(top)/SiO_x^(bottom) bilayer (with or without corner attachments) is deposited through PECVD and patterned into a U-shape with corner compensation through wet etching. (e) A photoresist cover is then added to the roots of the U-shape legs. (f) The desired rolled-up microtube to be achieved after etching off the sacrificial layer.

To further validate the effectiveness of corner engineering in practice, microfabrication experiments based on rolled-up nanotechnology were performed to compare the success rate (roll-up yield) of the nanomembrane roll-up process with or without corner attachments. A one end clamped U-shape nanomembrane, like the one used in FE simulations was utilized in the experimental design. The bilayer nanomembranes consist of the bottom SiO_{x'} layer and the top SiN_x layer, and both were synthesised by plasma enhanced chemical vapor deposition (PECVD). The fully-automated PECVD machine allows fine tuning of the gas ratio and working frequency for thin-film deposition, through which the stress difference between the two layers can be reproducibly controlled (compressive stress in bottom SiOx' layer and tensile stress in upper SiN_x layer) for generating enough stress difference to enable the rolling process. The dimensions of the U-shape nanomembrane are shown in Fig. 4.8d, with the corner attachment of 20 μ m in length and 5 μ m in width. As indicated in the predicted rolling direction mapping in Fig. 4.8b, this length/width ratio of the corner attachment is expected to lead to longitude roll-up. For fabricating the rolled-up microtube, an Al sacrificial layer (50 nm) was first deposited on a cleaned three-inch silicon wafer using E-beam deposition (BJD 1800, Temescal) (Fig. 4.8c). A SiN_x, (top)/SiO_x (bottom) bilayer was then deposited by PECVD (Plasmalab System100, Oxford Instruments) with deposition parameters of 8.5 sccm SiH₄ with 710 sccm N₂O, and 10 sccm SiH₄ with 10 sccm NH₃ for SiO_x and SiN_{x'}, respectively. The bilayer was patterned into U-shapes with and without additional corners (Fig.4.8d) through hydrofluoric acid wet etching (all other parameters remained the same). A rectangular photoresist strip was then patterned to cover the roots of the U-shape legs, which was used to anchor the rolled-up tubes during the releasing process (Fig. 4.8e). The leg cover also defined the length to width ratio the U-shape for the subsequent releasing step, as shown in Fig. 4.8e. Aluminum etchant type A was used to remove the Al sacrificial layer for releasing the bilayer from its substrate (Fig. 4.8f), and the released samples were finally dried in a CO₂ critical point dryer (Tousimis). As shown in Figs 4.9a and b, the roll-up yield of the U-shape nanomembrane with additional corners was significantly improved. The rolling-up yield of each design (with or without additional corners)

is quantified based on the statistics from three fabrication batches, with 100 U-shape patterns in each batch. The result was shown in Fig. 4.9c, and the rolling-up yield was improved from $10.0\pm1.63\%$ to $87.7\pm2.49\%$ (*n*=3 batches). The experiment results provide solid evidence supporting the theory predication that by adding the additional corners, the transverse roll-up could be suppressed to allow short-edge (i.e., longitude) rolling to dominate the roll-up process. Basically, the additional corner locally alters the length/width ratio the U-shape nanomembrane, effectively rendering the original short-edge to become the long-edge. This demonstration illustrates the usefulness of the proposed theory for guiding practical microtube fabrication experiments. We believe that the theory will provide useful guidelines for designing the nanomembrane geometry and enhancing the roll-up yield.



Figure 4.9. SEM photographs for the microtubes fabricated (a) with and (b) without additional corners, along with the corresponding (c) roll-up yield (n = 3 batches, 100 tubes/batch) for the two cases. The insets on the upper right of (a) and (b) indicate the enlarged single microtubes obtained from experiments, compared with the ones obtained from FE simulation. The scale bars in the enlarged images indicate 30 µm.

4.5 Conclusion

In summary, this work presented a quantitative theoretical model for systematic investigation of the self-rolling of nanomembranes under different lattice mismatch strain anisotropy. The effects of lattice mismatch strain anisotropy on multiple equilibrium states of the rolled-up nanomembrane have been investigated. Analytical expressions have been obtained for predicting the roll-up curvatures, validated through finite element (FE) simulations. The present study explicitly demonstrates the bidirectional nature of the self-rolling of nanomembranes, and clarifies the critical role of transverse strain in contributing to the discrepancy between previous model predictions and experimental measurements. On base of our theoretical results, a couple of fabrication strategies, i.e., third-layer deposition and corner geometry engineering, have been proposed to predictively manipulate the bidirectional rolling competition of strained nanomembranes, to achieve controlled, unidirectional roll-up. In particular, for the strategy of corner engineering, its practical applications have been verified through microfabrication experiments, demonstrating great effectiveness in improving the roll-up yield of microtubes. Our work will provide useful guidelines for design and prediction of novel 3D nanomembrane geometries and suggest significant potential for optimization of the nanomembrane roll-up yield.

4.6 Acknowledgement

We greatly thank the financial support from McGill Engineering Doctoral Award, China Scholarship Council and Natural Sciences and Engineering Research Council grant (Grant # STPGP 463182-14). We also acknowledge Supercomputer Consortium Laval UQAM McGill and Eastern Quebec for providing computing power.

4.7 Supporting information

4.7.1 Derivation details for analytical formulation in Section 4.3.1

Below we elaborate on the derivation of those equations in Section 4. 2.1 in the manuscript. The strain components within the top and bottom layers of a rectangular bilayer can be expressed as

$$\begin{bmatrix} \varepsilon_{11}^{t} \\ \varepsilon_{12}^{t} \\ \varepsilon_{22}^{t} \end{bmatrix} = \begin{bmatrix} 3C_{2}x_{1}^{2} + C_{3}x_{2}^{2} + C_{1} + \frac{1}{2}\kappa_{11}^{2}x_{1}^{2} + x_{3}\kappa_{11} - \varepsilon_{0} \\ 3C_{5}x_{2}^{2} + C_{6}x_{1}^{2} + C_{4} + \frac{1}{2}\kappa_{22}^{2}x_{2}^{2} + x_{3}\kappa_{22} \\ C_{3}x_{1}x_{2} + C_{6}x_{1}x_{2} + \frac{1}{2}\kappa_{11}\kappa_{22}x_{1}x_{2} - \eta \cdot \varepsilon_{0} \end{bmatrix}$$
(4.19)

$$\begin{bmatrix} \varepsilon_{11}^{b} \\ \varepsilon_{12}^{b} \\ \varepsilon_{22}^{b} \end{bmatrix} = \begin{bmatrix} 3C_{2}x_{1}^{2} + C_{3}x_{2}^{2} + C_{1} + \frac{1}{2}\kappa_{11}^{2}x_{1}^{2} + x_{3}\kappa_{11} \\ 3C_{5}x_{2}^{2} + C_{6}x_{1}^{2} + C_{4} + \frac{1}{2}\kappa_{22}^{2}x_{2}^{2} + x_{3}\kappa_{22} \\ C_{3}x_{1}x_{2} + C_{6}x_{1}x_{2} + \frac{1}{2}\kappa_{11}\kappa_{22}x_{1}x_{2} \end{bmatrix}$$
(4.20)

Assuming the state of plane stress for the rectangular nanomembrane, the stress components of substrate and film ($\sigma_{ij}^b, \sigma_{ij}^t$) could be calculated based on the strain-stress relationship

$$\sigma_{\alpha\beta} = \frac{E}{(1+\nu)} \Big(\varepsilon_{\alpha\beta} + \frac{\nu}{(1-\nu)} \varepsilon_{\gamma\gamma} \delta_{\alpha\beta} \Big)$$
(4.21)

The total strain energy of the system follows by integrating the strain energy over the volume as

$$U = \iiint_V \left(\frac{1}{2}\sigma^b_{ij}\varepsilon^b_{ij} + \frac{1}{2}\sigma^t_{ij}\varepsilon^t_{ij}\right)dV$$
(4.22)

The coefficients and curvatures can now be determined by minimizing the potential energy U. For simplicity, assuming $v_b = v_t = v$, $\eta = 1$ and W = L, we can obtain the analytical solutions of coefficients C_i in terms of the mismatch strain and curvature by solving the six simultaneous equations $\partial U/\partial C_i=0$:

$$C_{1} = \begin{pmatrix} \frac{1}{12} \frac{\left(\left(\left((6\nu+36)h_{t}-5L^{2}k_{22}(\nu-1)\right)E_{b}-6E_{t}h_{t}(\nu+6)\right)k_{11}h_{b}\right)}{(E_{b}h_{b}+E_{t}h_{t})(\nu+6)} \\ -\frac{1}{12} \frac{5L^{2}E_{t}h_{t}k_{11}k_{22}(\nu-1)}{(E_{b}h_{b}+E_{t}h_{t})(\nu+6)} \\ +\frac{E_{b}h_{b}\varepsilon_{0}}{(E_{b}h_{b}+E_{t}h_{t})} \end{pmatrix}$$
(4.23)

$$C_2 = \frac{(2(\nu+6)k_{11} - 5k_{22}\nu)k_{11}}{12\nu + 72} \tag{4.24}$$

$$C_3 = \frac{5k_{11}k_{22}}{4\nu + 24} \tag{4.25}$$

$$C_{4} = \begin{pmatrix} \frac{1}{12} \frac{\left(\left(\left((6\nu+36)h_{t} - 5L^{2}k_{11}(\nu-1) \right)E_{b} - 6E_{t}h_{t}(\nu+6) \right)k_{22}h_{b} \right)}{(E_{b}h_{b} + E_{t}h_{t})(\nu+6)} \\ - \frac{1}{12} \frac{5L^{2}E_{t}h_{t}k_{11}k_{22}(\nu-1)}{(E_{b}h_{b} + E_{t}h_{t})(\nu+6)} \\ + \frac{E_{b}h_{b}\varepsilon_{0}}{(E_{b}h_{b} + E_{t}h_{t})} \end{pmatrix}$$
(4.26)

$$C_5 = \frac{(2(\nu+6)k_{22}-5k_{11}\nu)k_{22}}{12\nu+72} \tag{4.27}$$

$$C_6 = \frac{5k_{11}k_{22}}{4\nu + 24} \tag{4.28}$$

Substituting the results of C_i back into the expression of U, and using the two remaining conditions $\partial U/\partial \kappa_{11} = 0$, $\partial U/\partial \kappa_{22} = 0$ yields the equilibrium equations (10-11) of κ_{11} and κ_{22} .

$$\begin{pmatrix} +E_b^2 h_b^2 \left(-\frac{3}{2} (\nu+6)(k_{22}\nu+k_{11})h_b^2 + L^4 k_{11}k_{22}^2 (\nu-1)(\nu+1) \right) \\ -3(\nu+6)(k_{22}\nu+k_{11})h_t^2 \\ -\frac{9}{2} \left((k_{22}\nu+k_{11})h_s + \varepsilon_0(\nu+1) \right)(\nu+6)h_b \\ -3(\nu+6)(k_{22}\nu+k_{11})h_b^2 \\ -\frac{9}{2} \varepsilon_0(\nu+6)(\nu+1)h_b \\ +L^4 k_{11}k_{22}^2 (\nu-1)(\nu+1) \end{pmatrix} \\ +h_t^2 E_t^2 \left(-\frac{3}{2} (\nu+6)(k_{22}\nu+k_{11})h_t^2 + L^4 k_{11}k_{22}^2 (\nu-1)(\nu+1) \right) \end{pmatrix} = 0$$

$$(4.29)$$

$$\begin{pmatrix} +h_b^2 E_b^2 \left(-\frac{3}{2} (\nu+6)(k_{11}\nu+k_{22})h_b^2 + L^4 k_{11}^2 k_{22} (\nu-1)(\nu+1) \right) \\ -3(\nu+6)(k_{11}\nu+k_{22})h_t^2 \\ -\frac{9}{2} (\nu+6) \left((k_{11}\nu+k_{22})h_b + \varepsilon_0(\nu+1) \right) h_t \\ -3(\nu+6)(k_{11}\nu+k_{22})h_b^2 \\ -\frac{9}{2} \varepsilon_0(\nu+6)(\nu+1)h_b \\ +L^4 k_{11}^2 k_{22} (\nu-1)(\nu+1) \end{pmatrix} \right) = 0$$

$$(4.30)$$

A closed-form relationship between longitudinal and transverse curvatures can then be obtained:

$$(\kappa_{11} - \kappa_{22}) \left(\kappa_{11} \kappa_{22} - \frac{3}{2} \frac{(\nu+6) \left(h_t^4 E_t^2 + 4h_t \left(h_t^2 + \frac{3}{2} h_b h_t + h_b^4 \right) E_b E_t h_b + h_b^4 E_b^2 \right)}{L^4 (E_b h_b + E_t h_t)^2 (\nu+1)} \right) = 0$$
(4.31)

Eq. 13 indicates that multiple equilibrium states for the bidirectional roll-up exist, as illustrated in **Figs. 1b-c**, in the two categories of (I) $\kappa_{11} = \kappa_{22}$, and (II) the two curvatures being related as

$$\kappa_{11}\kappa_{22} = \frac{3}{2} \frac{(\nu+6)\left(h_t^4 E_t^2 + 4h_t\left(h_t^2 + \frac{3}{2}h_bh_t + h_b^4\right)E_bE_th_b + h_b^4E_b^2\right)}{L^4(E_bh_b + E_th_t)^2(\nu+1)}$$
(4.32)

The explicit expressions of curvatures can be obtained if we further assume that the two layers are of the same thickness and elastic modulus, i.e., $h_t = h_b = h$, and $E_b = E_t$ yielding:

$$-6(\nu+6)(k_{22}\nu+k_{11})h^2 - \frac{9}{2}\varepsilon_0(\nu+6)(\nu+1)h + L^4k_{11}k_{22}^2(\nu-1)(\nu+1) = 0 \quad (4.33)$$

$$-6(\nu+6)(k_{11}\nu+k_{22})h^2 - \frac{9}{2}\varepsilon_0(\nu+6)(\nu+1)h + L^4k_{22}k_{11}^2(\nu-1)(\nu+1) = 0 \quad (4.34)$$

Category (I):

$$\kappa_{11} = \kappa_{22} = \frac{2^{4/3} \left(\frac{1}{4} (h(\nu+6) (9L^2 \varepsilon_0 + A) (\nu-1)^2)^{2/3} + (\nu^2 + 5\nu - 6) h^2 2^{1/3} \right)}{(h(\nu+6) (9L^2 \varepsilon_0 + A) (\nu-1)^2)^{1/3} L^2 (\nu-1)},$$
(4.35)

where
$$A = \sqrt{\frac{81L^4(\nu-1)\varepsilon_0^2 - 128h^4(\nu+6)}{\nu-1}}$$
.

Category (II):

$$(\kappa_{11}, \kappa_{22}) = (\kappa^+, \kappa^-) \text{ or } (\kappa_{11}, \kappa_{22}) = (\kappa^-, \kappa^+)$$
 (4.36)

with

$$\kappa^{+} = \frac{\sqrt{3}}{8} \frac{\sqrt{\frac{3L^{4}(\nu+1)^{3}\varepsilon_{0}^{2} - 128h^{4}(\nu+6)}{\nu+1}} + 3\varepsilon_{0}(\nu+1)L^{2}}{L^{2}h}$$
(4.37)

$$\kappa^{-} = \frac{\sqrt{3}}{8} \frac{\sqrt{\frac{3L^{4}(\nu+1)^{3}\varepsilon_{0}^{2} - 128h^{4}(\nu+6)}{\nu+1}} - 3\varepsilon_{0}(\nu+1)L^{2}}{L^{2}h}$$
(4.38)

From Eqs. 4.37-38, we can see that in the particular case of very thin film, i.e., $h \ll L$ (and W), the curvature $\kappa^- \to 0$, being negligible compared to κ^+ , and consequently the roll-up effectively becomes unidirectional. Therefore, assuming $\kappa_{11} = 0$ or $\kappa_{22} = 0$ for the $\partial U/\partial \kappa_{11} = 0$, $\partial U/\partial \kappa_{22} = 0$, respectively, we can obtain explicit solutions of curvatures in more general conditions ($v_b = v_t = v$, and W = L) as the following with $\rho = \frac{h_t}{h_b}$ and $E_r = \frac{E_t}{E_b}$

$$\kappa_{22} = 0, \ \kappa_{11} = \frac{6 \cdot \varepsilon_0 \cdot E_r \cdot \rho \cdot (\rho + 1)(1 + \eta \nu)}{h_b \left(\rho^4 + 4\rho \cdot \left(\rho^2 + \frac{3}{2}\rho + 1\right)E_r + E_r^2\right)}$$
(4.39)

$$\kappa_{11} = 0, \ \kappa_{22} = \frac{6 \cdot \varepsilon_0 \cdot E_r \cdot \rho \cdot (\rho + 1)(\nu + \eta)}{h_b \left(\rho^4 + 4\rho \cdot \left(\rho^2 + \frac{3}{2}\rho + 1\right)E_r + E_r^2\right)}$$
(4.40)

4.7.2 Derivation details for the third layer deposition in Section 4.4.2

The effects of third-layer deposition on roll-up are examined following a similar process carried out for the bilayer system. Here, we denote the Young's modulus, thicknesses, and Poisson's ratio of the third layer as E_3 , h_3 , and v_3 . For simplicity but without loss of generality, it is assumed that the third-layer results in an isotropic mismatch strain ε_1 and that the three layers exhibit the same Poisson's ratio and Young's modulus, $v_b = v_t = v_3 = v$ and $E_b = E_t = E_3 = E$, with a size of W = L. The total strain energy of the system is obtained by integrating the strain energy of three layers over the whole volume.

$$U = \iiint_V \left(\frac{1}{2}\sigma_{ij}^b \varepsilon_{ij}^b + \frac{1}{2}\sigma_{ij}^t \varepsilon_{ij}^t + \frac{1}{2}\sigma_{ij}^3 \varepsilon_{ij}^3\right) dV$$
(4.41)

Where, σ_{ij}^3 and ε_{ij}^3 are the stress and strain components of the third layer. The similar procedure has been conducted to get two equations determining κ_{11} and κ_{22} .

$$\begin{pmatrix} \left(\frac{1}{6}h_3^3 + \frac{1}{2}(h_t + h_b)h_3^2 + \left(\frac{1}{2}h_t^2 + h_th_b + \frac{1}{2}h_b^2\right)h_3 \\ + \frac{1}{6}h_t^3 + \frac{1}{2}h_t^2h_b + \frac{1}{2}h_b^2h_t + \frac{1}{6}h_b^3 \end{pmatrix}k_{22} + \\ \left(\left(\varepsilon_1(\nu+1) - \varepsilon_0(\nu+\eta)\right)h_t + \varepsilon_1h_b(\nu+1)\right)h_3 + \varepsilon_0h_th_b(\nu+\eta) \end{pmatrix} = 0$$
(4.42)

$$\begin{pmatrix} \left(\frac{1}{6}h_{3}^{3} + \frac{1}{2}(h_{t} + h_{b})h_{3}^{2} + \left(\frac{1}{2}h_{t}^{2} + h_{t}h_{b} + \frac{1}{2}h_{b}^{2}\right)h_{3} \\ + \frac{1}{6}h_{t}^{3} + \frac{1}{2}h_{t}^{2}h_{b} + \frac{1}{2}h_{b}^{2}h_{t} + \frac{1}{6}h_{b}^{3} \end{pmatrix}k_{11} \\ + \left((\eta\varepsilon_{0}\nu + \varepsilon_{1}\nu + \varepsilon_{1} - \varepsilon_{0})h_{t} + \varepsilon_{1}h_{b}(\nu + 1)\right)h_{3} \\ + (\eta\nu + 1)\varepsilon_{0}h_{t}h_{b} \end{pmatrix}$$
(4.43)

Then we can obtain solutions of curvatures for longitude roll-up and transverse roll-up:

Longitude roll-up:

$$\kappa_{22} = 0, \kappa_{11} = \frac{\left((6\eta\varepsilon_1\nu - 6\varepsilon_0\nu - 6\varepsilon_0 + 6\varepsilon_1)h_t - 6\varepsilon_1h_bh_t(\eta\nu + 1)\right) - 6\varepsilon_0h_3h_s(\nu + 1)}{(h_3 + h_t + h_b)^3}$$
(4.44)

Transverse roll-up:

$$\kappa_{11} = 0, \kappa_{22} = \frac{\left((6(\eta+\nu)\varepsilon_1 - (6\nu+6)\varepsilon_0)h_t - 6\varepsilon_1h_bh_t(\eta+\nu)\right) - 6\varepsilon_0h_3h_b(\nu+1)}{(h_3 + h_t + h_b)^3}$$
(4.45)

Afterwards, from Eq. (27), we solve the h_3 that makes $\kappa_{22} = 0$, which is the critical third layer thickness h_3^c that eliminates the transverse effect;

$$h_3^c = -\frac{\varepsilon_0 h_t h_b(\nu+\eta)}{(h_t+h_b)(1+\nu)\varepsilon_0 - \varepsilon_1(\nu+\eta)h_t}$$
(4.46)

At $h_3 = h_3^c$, the rolling is only viable along the longitude direction, and the corresponding curvature is

$$\kappa_{11}^{c} = \frac{6(\nu^{2}-1)(((\eta+\nu)\varepsilon_{1}-\varepsilon_{0}(\nu+1))h_{t}-\varepsilon_{0}h_{b}(1+\nu))^{2}(\eta-1)h_{t}h_{b}(h_{t}+h_{b})\varepsilon_{1}\varepsilon_{0}}{(((\eta+\nu)\varepsilon_{1}-\varepsilon_{0}(\nu+1))h_{t}^{2}+2h_{b}((\eta+\nu)\varepsilon_{1}-\varepsilon_{0}(\nu+1))h_{t}-\varepsilon_{0}h_{b}^{2}(1+\nu))^{3}}$$
(4.47)

4.8 References

[1] C.a. Vervacke, Three-dimensional chemical sensors based on rolled-up hybrid nanomembranes, RSC Advances, 4 (2014) 9723.

[2] G.J. Wan, A.A. Solovev, G.S. Huang, M.F. Maitz, N. Huang, Y.F. Mei, Dynamic curvature control of rolled-up metal nanomembranes activated by magnesium, Journal of Materials Chemistry, 22 (2012) 12983-12987.

[3] R. Arayanarakool, A.K. Meyer, L. Helbig, S. Sanchez, O.G. Schmidt, Tailoring threedimensional architectures by rolled-up nanotechnology for mimicking microvasculatures, Lab Chip, 15 (2015) 2981-2989.

[4] I. Mnch, D. Makarov, R. Koseva, L. Baraban, D. Karnaushenko, C. Kaiser, K.F. Arndt, O.G. Schmidt, Rolled-up magnetic sensor: Nanomembrane architecture for in-flow detection of magnetic objects, ACS Nano, 5 (2011) 7436-7442.

[5] S.M. Harazim, V.A.B. Quinones, S. Kiravittaya, S. Sanchez, O.G. Schmidt, Lab-in-a-tube: on-chip integration of glass optofluidic ring resonators for label-free sensing applications, Lab Chip, 12 (2012) 2649-2655.

[6] a. Bernardi, S. Kiravittaya, a. Rastelli, R. Songmuang, D.J. Thurmer, M. Benyoucef, O.G. Schmidt, On-chip Si/SiO_x microtube refractometer, Appl Phys Lett, 93 (2008).

[7] S.M. Harazim, W. Xi, C.K. Schmidt, S. Sanchez, O.G. Schmidt, Fabrication and applications of large arrays of multifunctional rolled-up SiO/SiO2 microtubes, Journal of Materials Chemistry, 22 (2012) 2878.

[8] D.a. Grimm, Rolled-up nanomembranes as compact 3D architectures for field effect transistors and fluidic sensing applications, Nano Lett, 13 (2013) 213-218.

[9] O.G. Schmidt, C. Deneke, S. Kiravittaya, R. Songmuang, H. Heidemeyer, Y. Nakamura, R. Zapf-Gottwick, C. Muller, N.Y. Jin-Phillipp, Self-assembled nanoholes, lateral quantum-dot molecules, and rolled-up nanotubes, Ieee J Sel Top Quant, 8 (2002) 1025-1034.

[10] D.J. Bell, L. Dong, B.J. Nelson, M. Golling, Fabrication and Characterization of Three-Dimensional InGaAs GaAs Nanosprings, Nano, (2006).

[11] K. Malachowski, M. Jamal, Q. Jin, B. Polat, C.J. Morris, D.H. Gracias, Self-folding single cell grippers, Nano Lett, 14 (2014) 4164-4170.

[12] W.M. Li, G.S. Huang, J. Wang, Y. Yu, X.J. Wu, X.G. Cui, Y.F. Mei, Superelastic metal microsprings as fluidic sensors and actuators, Lab Chip, 12 (2012) 2322-2328.

[13] M.H. Huang, C. Boone, M. Roberts, D.E. Savage, M.G. Lagally, N. Shaji, H. Qin, R. Blick, J.A. Nairn, F. Liu, Nanomechanical architecture of strained bilayer thin films: From design principles to experimental fabrication, Adv Mater, 17 (2005) 2860-2864.

[14] Z. Ren, P.X. Gao, A review of helical nanostructures: growth theories, synthesis strategies and properties, Nanoscale, 6 (2014) 9366-9400.

[15] B.A. Korgel, Nanosprings take shape, Science, 309 (2005) 1683-1684.

[16] Y.F. Mei, G.S. Huang, A.A. Solovev, E.B. Urena, I. Moench, F. Ding, T. Reindl, R.K.Y. Fu, P.K. Chu, O.G. Schmidt, Versatile Approach for Integrative and Functionalized Tubes by Strain Engineering of Nanomembranes on Polymers, Adv Mater, 20 (2008) 4085-4090.

[17] I.S. Chun, V.B. Verma, V.C. Elarde, S.W. Kim, J.M. Zuo, J.J. Coleman, X. Li, InGaAs/GaAs 3D architecture formation by strain-induced self-rolling with lithographically defined rectangular stripe arrays, J Cryst Growth, 310 (2008) 2353-2358.

[18] X.L. Li, Strain induced semiconductor nanotubes: from formation process to device applications, J Phys D Appl Phys, 41 (2008).

[19] Y.V. Nastaushev, V.Y. Prinz, S.N. Svitasheva, A technique for fabricating Au/Ti microand nanotubes, Nanotechnology, 16 (2005) 908-912.

[20] P. Cendula, S. Kiravittaya, I. Monch, J. Schumann, O.G. Schmidt, Directional Roll-up of Nanomembranes Mediated by Wrinkling, Nano Lett, 11 (2011) 236-240.

[21] W. Huang, S. Koric, X. Yu, K.J. Hsia, X.L. Li, Precision Structural Engineering of Self-Rolled-up 3D Nanomembranes Guided by Transient Quasi-Static FEM Modeling, Nano Lett, 14 (2014) 6293-6297.

[22] Z. Chen, G.S. Huang, I. Trase, X.M. Han, Y.F. Mei, Mechanical Self-Assembly of a Strain-Engineered Flexible Layer: Wrinkling, Rolling, and Twisting, Phys Rev Appl, 5 (2016).

[23] T. Huang, Z.Q. Liu, G.S. Huang, R. Liu, Y.F. Mei, Grating-structured metallic microsprings, Nanoscale, 6 (2014) 9428-9435.

[24] B.C. Li, Q. Wang, E.Y. Wang, Z.G. Jia, Y.C. Yan, Z.Q. Bian, X.M. Ren, S.W. Cai, Y.Q. Huang, Dependence of doubly curved regions on drying method in the fabrication of long-side rolled-up III-V microtubes, Appl Phys Lett, 103 (2013).

[25] I.S. Chun, A. Challa, B. Derickson, K.J. Hsia, X.L. Li, Geometry Effect on the Strain-Induced Self-Rolling of Semiconductor Membranes, Nano Lett, 10 (2010) 3927-3932.

[26] Y.Y. Hu, W.M. Huang, Elastic and elastic-plastic analysis of multilayer thin films: Closed-form solutions, J Appl Phys, 96 (2004) 4154-4160.

[27] G.P. Nikishkov, Curvature estimation for multilayer hinged structures with initial strains, J Appl Phys, 94 (2003) 5333-5336.

[28] Y.C. Tsui, T.W. Clyne, An analytical model for predicting residual stresses in progressively deposited coatings .1. Planar geometry, Thin Solid Films, 306 (1997) 23-33.

[29] M. Grundmann, Nanoscroll formation from strained layer heterostructures, Appl Phys Lett, 83 (2003) 2444-2446.

[30] J. Zang, M.H. Huang, F. Liu, Mechanism for nanotube formation from self-bending nanofilms driven by atomic-scale surface-stress imbalance, Phys Rev Lett, 98 (2007).

[31] O. Schumacher, S. Mendach, H. Welsch, A. Schramm, C. Heyn, W. Hansen, Lithographically defined metal-semiconductor-hybrid nanoscrolls, Appl Phys Lett, 86 (2005).

[32] C. Deneke, C. Muller, N.Y. Jin-Phillipp, O.G. Schmidt, Diameter scalability of rolled-up In(Ga)As/GaAs nanotubes, Semicond Sci Tech, 17 (2002) 1278-1281.

[33] M.H. Huang, F. Cavallo, F. Liu, M.G. Lagally, Nanomechanical architecture of semiconductor nanomembranes, Nanoscale, 3 (2011) 96-120.

[34] I.S. Chun, X.L. Li, Controlled assembly and dispersion of strain-induced InGaAs/GaAs nanotubes, Ieee T Nanotechnol, 7 (2008) 493-495.

[35] G. Stoychev, S. Zakharchenko, S. Turcaud, J.W.C. Dunlop, L. Ionov, Shape-Programmed Folding of Stimuli-Responsive Polymer Bilayers, Acs Nano, 6 (2012) 3925-3934.

[36] E.V. Naumova, V.Y. Prinz, S.V. Golod, V.A. Seleznev, R.A. Soots, V.V. Kubarev, Manufacturing chiral electromagnetic metamaterials by directional rolling of strained heterofilms, J Opt a-Pure Appl Op, 11 (2009).

[37] A.B. Vorob'ev, V.Y. Prinz, Directional rolling of strained heterofilms, Semicond Sci Tech, 17 (2002) 614-616.

[38] K. Dietrich, C. Strelow, C. Schliehe, C. Heyn, A. Stemmann, S. Schwaiger, S. Mendach, A. Mews, H. Weller, D. Heitmann, T. Kipp, Optical Modes Excited by Evanescent-Wave-Coupled PbS Nanocrystals in Semiconductor Microtube Bottle Resonators, Nano Lett, 10 (2010) 627-631.

[39] T. Kipp, C. Strelow, D. Heitmann, Light Confinement in Microtubes, in: D. Heitmann (Ed.) Quantum Materials, Lateral Semiconductor Nanostructures, Hybrid Systems and Nanocrystals: Lateral Semiconductor Nanostructures, Hybrid Systems and Nanocrystals, Springer Berlin

Heidelberg, Berlin, Heidelberg, 2010, pp. 165-182.

[40] L.B. Freund, Substrate curvature due to thin film mismatch strain in the nonlinear deformation range, J Mech Phys Solids, 48 (2000) 1159-1174.

[41] S. Vidoli, Discrete approximations of the Foppl-Von Karman shell model: From coarse to more refined models, Int J Solids Struct, 50 (2013) 1241-1252.

[42] M.L. Dano, M.W. Hyer, Thermally-induced deformation behavior of unsymmetric laminates, Int J Solids Struct, 35 (1998) 2101-2120.

[43] M.W. Hyer, Calculations of the Room-Temperature Shapes of Unsymmetric Laminates, J Compos Mater, 15 (1981) 296-310.

[44] D.J. Bell, L.X. Dong, B.J. Nelson, M. Golling, L. Zhang, D. Grutzmacher, Fabrication and characterization of three-dimensional InGaAs/GaAs nanosprings, Nano Lett, 6 (2006) 725-729.
[45] F. Li, Z.T. Mi, Optically pumped rolled-up InGaAs/GaAs quantum dot microtube lasers, Optics Express, 17 (2009) 19933-19939.

[46] M.H.T. Dastjerdi, M. Djavid, S. Arafin, X. Liu, P. Bianucci, Z. Mi, P.J. Poole, Optically pumped rolled-up InAs/InGaAsP quantum dash lasers at room temperature, Semicond Sci Tech, 28 (2013).

[47] S. Timoshenko, Analysis of bi-metal thermostats, J Opt Soc Am Rev Sci, 11 (1925) 233-255.

[48] E.J. Barbero, Finite Element Analysis of Composite Materials using AbaqusTM, Taylor & Francis, 2013.

[49] P. Froeter, X. Yu, W. Huang, F. Du, M.Y. Li, I. Chun, S.H. Kim, K.J. Hsia, J.A. Rogers, X.L. Li, 3D hierarchical architectures based on self-rolled-up silicon nitride membranes, Nanotechnology, 24 (2013).

[50] L. Ionov, 3D Microfabrication using Stimuli-Responsive Self-Folding Polymer Films, Polym Rev, 53 (2013) 92-107.

[51] L. Zhang, S.V. Golod, E. Deckardt, V. Prinz, D. Grutzmacher, Free-standing Si/SiGe micro- and nano-objects, Physica E, 23 (2004) 280-284.

[52] R. Deki, T. Sasaki, M. Takahasi, Strain relaxation and compositional separation during growth of InGaAs/GaAs(001), J Cryst Growth, 468 (2017) 241-244.

[53] T. Mano, R. Notzel, G.J. Hamhuis, T.J. Eijkemans, J.H. Wolter, Direct imaging of selforganized anisotropic strain engineering for improved one-dimensional ordering of (In,Ga)As quantum dot arrays, J Appl Phys, 95 (2004) 109-114.

[54] I. Tangring, Y.X. Song, Z.H. Lai, S.M. Wang, M. Sadeghi, A. Larsson, A study of the doping influence on strain relaxation of graded composition InGaAs layers grown by molecular beam epitaxy, J Cryst Growth, 311 (2009) 1684-1687.

[55] O. Yastrubchak, T. Wosinski, J.Z. Domagata, E. Lusakowska, T. Figielski, B. Pecz, A.L. Toth, Misfit strain anisotropy in partially relaxed lattice-mismatched InGaAs/GaAs heterostructures, J Phys-Condens Mat, 16 (2004) S1-S8.

[56] J. Wang, E.M. Song, C.L. Yang, L.R. Zheng, Y.F. Mei, Fabrication and whispering gallery resonance of self-rolled up gallium nitride microcavities, Thin Solid Films, 627 (2017) 77-81.

[57] Z. Tian, L. Zhang, Y.F. Fang, B. Xu, S.W. Tang, N. Hu, Z.H. An, C. Zi, Y.F. Mei, Deterministic Self-Rolling of Ultrathin Nanocrystalline Diamond Nanomembranes for 3D Tubular/Helical Architecture, Adv Mater, 29 (2017).

[58] S. Alben, B. Balakrisnan, E. Smela, Edge Effects Determine the Direction of Bilayer Bending, Nano Lett, 11 (2011) 2280-2285.

[59] S.L. Li, L.B. Ma, S. Bottner, Y.F. Mei, M.R. Jorgensen, S. Kiravittaya, O.G. Schmidt, Angular position detection of single nanoparticles on rolled-up optical microcavities with lifted degeneracy, Phys Rev A, 88 (2013).

[60] F.Y. Zhao, T.R. Zhan, G.S. Huang, Y.F. Mei, X.H. Hu, Liquid sensing capability of rolledup tubular optical microcavities: a theoretical study, Lab Chip, 12 (2012) 3798-3802.

[61] J. Wang, T.R. Zhan, G.S. Huang, P.K. Chu, Y.F. Mei, Optical microcavities with tubular geometry: properties and applications, Laser Photonics Rev, 8 (2014) 521-547.

[62] P. Bianucci, S. Mukherjee, M.H.T. Dastjerdi, P.J. Poole, Z.T. Mi, Self-organized InAs/InGaAsP quantum dot tube lasers, Appl Phys Lett, 101 (2012).

Chapter 5:Topological patterning effects on the self-rolling of nanomembranes

As an important "top down" nanofabrication approach, surface patterning through photolithography is generally combined with the self-rolling technique of strained engineered nanomembranes to guide the deposition or removal of material achieving a higher level of manipulation over the rolling process for creating various sophisticated non-planar rollup nanostructures, such as nanotubes, helix, and wrinkles. In this chapter, we develop an analytical model aiming at examining the effects of patterning on self-rolling behaviors, where the patterning induced local film thickness variation are effectively accounted for based on Von-Karman nonlinear theory and Ritz method. The dependence of rollup direction and curvature of a nanomembrane on the size and density of patterns has been systematically studied. The accuracy of our theoretical model has been demonstrated by its application to helix-shape structures to showcase a quantitative route to realize controllable helical rolling of nanomembranes through strip patterning, showing excellent agreement with both experimental observations and finite-element simulations. Our results provide critical insights for the design of complex 3D structures of self-assembly of nanomembranes through controllable surface patterning.

• This chapter has been published in *Nanotechnology*, appeared as:

Topological patterning effects on the self-rolling of nanomembranes, *Nanotechnology* 29 (2018), 345301.

Cheng Chen, Pengfei Song, Fanchao Meng, Pengfei Ou, Xinyu Liu, Jun Song*

5.1 Abstract

The effects of topological patterning (i.e., grating and rectangular patterns) on the selfrolling behaviors of heteroepitaxial strained nanomembranes have been systematically studied. An analytical modeling framework, validated through finite-element simulations, has been formulated to predict the resultant curvature of the patterned nanomembrane as the pattern thickness and density vary. The effectiveness of the grating pattern in regulating the rolling direction of the nanomembrane has been demonstrated and quantitatively assessed. Further to the rolling of nanomembranes, a route to achieve predictive design of helical structures has been proposed and showcased. The present study provides new knowledge and mechanistic guidance towards predictive control and tuning of roll-up nanostructures via topological patterning.

5.2 Introduction

With continuous progression towards device and component miniaturization in nanotechnology, there is increasing demand in design flexibility and geometry variability [1-3]. Self-rolling of nanomembranes, where a strained bilayer (or multilayer) thin film scrolls up to form a three-dimensional (3D) structure, provides a powerful fabrication route towards realizing various 3D nanoscale configurations, such as nanotubes [4, 5], helix [6], and wrinkles [7, 8]. These unique 3D structures, combing with plentiful pool of available functional materials, has motivated numerous applications in nanotechnology, such as microscale resonators [9-11], smart energy storage devices [12-14], and micro engines [15, 16], where the roll-up nanostructures are employed as primary architecture units, promoting great technological progress in micro/nano-electro-mechanical systems (MEMS/NEMS) and lab on a chip [17-20].

However, the fabrication and obtainment of those sophisticated non-planar roll-up nanostructures are heavily dependent on the topographic patterning technique that predefine the initial planar geometry to guide the deposition or removal of material of the strain engineered nanomembrane in order to achieve a higher level of manipulation over the rolling process [21-24]. For instance, the patterned grating structures have proved to be an effective strategy that can define and control the rolling direction of strained nanomembranes to facilitate the fabrication of complex structure of micro-/nano-springs [25]. The patterned U-shape nanomembranes are necessary in microtube optical sensors for lifting-up the middle part of the microtube from the substrate after rolling-up [9, 26]. Meanwhile, the topographic patterning also plays an important role in the integration of state-of-the-art functionality with well-designed optical or electrical modules, providing exceptional design and fabrication flexibilities into targeted devices, thus enhancing the performance of existing structures and devices [27, 28].

The accurate design and fabrication of sophisticated roll-up configurations with patterns necessitate in-depth understanding of the mechanics underlying the rolling-up. Many analytical and numerical methods [29-34] have been developed, attempting to quantitatively simulate the roll-up process and to determine the resultant roll-up curvature [35-43]. However, in those previous modeling efforts, the effects of local thickness variation and nonuniform distribution of mismatch strain induced by patterning have not been considered.

The present study aims to address the afore-mentioned deficit in the modeling and simulation of self-rolling of patterned nanomembranes. Based on Von-Karman shell theory [44, 45] and Ritz method [46, 47], a comprehensive analytical model has been formulated. The new model quantitatively accounts for the effect of topographic patterning on the rolling direction and curvature of rolled-up nanomembranes. The accuracy and reliability of the analytical prediction from our model have been confirmed by finite-element (FE) simulations. Furthermore, the implication of our results to the design of more complex 3D roll-up structures (e.g., helical structures) was discussed, and a strategy to design helixes of well controlled geometric metrics, from a patterned host nanomembrane was demonstrated.

5.3 Methodology

5.3.1 Analytical formulation

Figure 5.1 illustrates the rectangular nanomembrane model considered in the present study. The length and width of the nanomembrane are denoted as L and W, with two axes, x_1 and x_2 , indicating the length and width directions respectively, also referred to as longitude (x_1) and lateral (x_2) directions below. The nanomembrane system comprises of two principle material layers, a passive substrate and an active film, denoted below as top and bottom layers respectively. On top of these two layers resides an additional pattern layer that may vary in size, density and thickness. The top layer with Young's modulus E_t , Poisson's ratio v_t , and
thicknesses h_t , is subject to initial mismatch strain ε_0 that originates from lattice mismatch or thermal loading. The elastic properties of the bottom layer of thickness h_b are characterized by Young's modulus E_b and Poisson's ratio v_b . Without loss of generality, the patterned material is assumed to be the same as the top layer with thickness h_p . In particular, the patterns can be divided into two categories, i.e., attached and etched patterns. These two categories can be selfconsistently characterized as $h_p > 0$ (attached) and $h_p < 0$ (etched). Here for ease of modeling we also define a dimensionless thickness parameter, $H_p = h_p/h_t$, to reflect the pattern induced relative thickness change for the top layer. Regarding the pattern layer, here we consider two typical patterns, i.e., grating and rectangular patterns, and for each case, we assume the pattern to be uniform in size, density and thickness, and that the width of the grating pattern or length/width of the cubic pattern are much smaller than the resultant roll-up radius of curvature, for simplicity. Aiming at quantitatively evaluating the size effect of patterns, two parameters (ρ_1, ρ_2) are used to indicate the area density of the pattern layer. The density parameters ρ_1 and ρ_2 are defined as the ratios between the patterned area and normal area within the patterncontaining *unit* strips along x_1 and x_2 directions respectively (see Fig. 5.1c). It is apparent that when both ρ_1 and ρ_2 are finite (see Fig. 5.1d), the cubic patterns are achieved, otherwise we obtain the grating patterns (see Figs. 5.1e-f).



Figure 5.1. Schematic illustration of different pattern structures examined for the bilayer nanomembranes: (a) grating patterns colored in orange, (b) attached and etched cubic patterns, colored in orange and pink respectively. (c) Definition of the two pattern density parameters, ρ_1 and ρ_2 , with three representative examples of different pattern types/densities provided, as shown in (d).

In our calculations, the relations of strain ε and displacement field u in a deformed nanomembrane in a given coordinate (x_1, x_2, x_3) (cf. Fig. 5.1a) are determined based on the Von-Karman nonlinear theory [48, 49] describing the large transverse deflections of thin shell, as shown below,

$$\varepsilon_{\alpha\beta} = \frac{1}{2} \left(\frac{\partial u_{\alpha}}{\partial x_{\beta}} + \frac{\partial u_{\beta}}{\partial x_{\alpha}} + \frac{\partial u_{3}}{\partial x_{\beta}} \cdot \frac{\partial u_{3}}{\partial x_{\alpha}} \right) - x_{3} \frac{\partial^{2} u_{3}}{\partial x_{\alpha} \partial x_{\beta}}, \qquad (5.1)$$

with the stress σ is related to strain through Young's modulus *E* and Poisson's ratio *v* in the following compact form:

$$\sigma_{\alpha\beta} = \frac{E}{(1+\nu)} \Big(\varepsilon_{\alpha\beta} + \frac{\nu}{(1-\nu)} \varepsilon_{\gamma\gamma} \delta_{\alpha\beta} \Big) - \frac{E\varepsilon_0}{(1-\nu)} \delta_{\alpha\beta}, \tag{5.2}$$

where the Greek subscripts α , β and $\gamma=1$, 2, and ε_0 denotes the isotropic inelastic mismatch strain. The displacement field \boldsymbol{u} , and subsequently the stress and strain fields can be obtained by minimizing the total strain energy $U = \iiint_V \frac{1}{2} \sigma_{ij} \varepsilon_{ij} dV$, where *V* represents the total volume of the strained nanomembrane. The existence of patterns, however, renders the surface nonuniform, in which case the strain energy can be divided into two parts:

$$U = U_n + U_p \tag{5.3}$$

with

$$U_n = \frac{1}{2} \int_0^{h_t + h_b} dx_3 \iint_{\boldsymbol{\Omega}_{S_n}} \left(\sigma_{ij}^b \varepsilon_{ij}^b + \sigma_{ij}^t \varepsilon_{ij}^t \right) d\boldsymbol{\Omega}_{S_n}$$
(5.4)

$$U_p = \frac{1}{2} \int_0^{h_b + h_t + h_p} dx_3 \iint_{\boldsymbol{\Omega}_{S_p}} \left(\sigma_{ij}^b \varepsilon_{ij}^b + \sigma_{ij}^t \varepsilon_{ij}^t + \sigma_{ij}^p \varepsilon_{ij}^p \right) d\boldsymbol{\Omega}_{S_p}$$
(5.5)

where U_n and U_p represent the strain energies associated with the normal (pattern-free) area $\mathbf{\Omega}_{S_n}$ and patterned area $\mathbf{\Omega}_{S_p}$ respectively. The superscripts *b*, *t* and *p* in the stress and strain terms indicate their association with bottom, top and pattern layers respectively. Subsequently the equilibrium configurations of nanomembranes with specific patterns could be obtained via minimization of *U* based on the trial in-plane and out-of-plane displacement shape functions. An approximated displacement field, i.e., u_i (*i*=1, 2, 3) of the mid-plane of the nanomembrane can be assumed to be [45-48]:

$$u_1(x_1, x_2) = \sum_{i=0}^3 \sum_{j=0}^3 A_{ij} x_1^i x_2^j , \qquad (5.6)$$

$$u_2(x_1, x_2) = \sum_{i=0}^3 \sum_{j=0}^3 B_{ij} x_1^i x_2^j , \qquad (5.7)$$

$$u_3 = \frac{1}{2}(ax_1^2 + bx_2^2), \qquad (5.8)$$

where A_{ij} , B_{ij} , a, and b are adjustable parameters to be determined through the minimization of U. The above three-order polynomial approximation of the in-plane displacement filed was proved to be sufficient to provide an accurate description for analyzing the deformation behaviors of thin film and shell structures [50, 51]. The curvature fields of the deformed nanomembrane can then be determined as:

$$\kappa_{11} = \frac{\partial^2 u_3}{\partial x_1^2} \quad \text{and} \quad \kappa_{22} = \frac{\partial^2 u_3}{\partial x_2^2}$$
(5.9)

5.3.2 Numerical simulations

Finite element (FE) simulations were employed for direct numerical simulations of the self-rolling behaviors of nanomembranes and validation of the analytical prediction of the resultant roll-up curvatures. In this study, FE simulations were performed using the ABAQUS Unified FE software [52]. The layered structures with predefined patterns were created in the module of composite layups, and 4-node doubly curved thin/thick shell elements with reduced integration (S4R) were used to discretize the domain. The initial mismatch strain was achieved by assigning different thermal expansion coefficients to different layers and varying the temperature. Two categories of FE simulations were performed to examine the self-rolling behaviors of patterned nanomembranes, detailed as the follows:

1) <u>*Category I*</u>: For FE simulations in this category, the rolling process was examined using the static analysis process with large rotation considered. The direction of rolling is pre-defined with a moving boundary condition set up to achieve controlled release of the nanomembrane, where the model geometry is partitioned into ribbon segments (perpendicular to the rolling direction) and fixed initially, and then released gradually [53];

2) <u>*Category II*</u>: ABAQUS explicit/dynamic package was employed for simulations in this category to investigate the competition between rollup behaviours along different roll-up directions of strained nanomembranes. Unlike those static simulations in category I, no pre-

constrain was imposed to influence the roll-up direction of the nanomembrane, corresponding to the situation of isotropic etching release. Simulations in this category focus on strained nanomembranes with grating patterns, examining the role of geometrical anisotropic distribution of grating pattern structures in inducing anisotropic rolling mode and the formation of helical shapes. It is worth to note that the effect of cubic patterns on the rollup competition was not considered here due to their geometrical isotropy.

5.4 Results

5.4.1 Effect of patterning on roll-up curvature for unidirectional rolling

Two nanomembrane systems, i.e., GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom) and GaN^(top)/AIN^(bottom), which are widely used to fabricate self-assembly semiconductor nanostructures with various geometries [6, 41, 54, 55], are selected as representative systems. The superscripts '*top*' and '*bottom*' are used to respectively indicate the materials of the top and bottom layers within the nanomembranes. The corresponding material properties are listed in Table I. The FE simulated evolutions of the roll-up curvature κ_{11} , for controlled unidirectional rolling along the longitude (x_1) direction, as the dimensionless thickness parameter H_p (= h_p/h_t) and pattern density parameters (ρ_1, ρ_2) vary are presented in Figs 5.2 and 5.3 for GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom) and GaN^(top)/AlN^(bottom) nanomembrane systems respectively. As previously described, negative and positive values of H_p respectively indicate etched and attached patterns. As observed in Figs 5.2 and 5.3, there is excellent agreement between the FE simulated and analytically predicted (from Eqs (5. 6-9)), which evidences the accuracy and reliability of the analytical model in describing the rolling behaviors of different material systems.

We note that for positive H_p , the increment in pattern density leads to decrease in the longitude rolling curvature κ_{11} , while the opposite dependence of curvature on pattern density

is observed for the negative range of H_p . It can also be seen that the influence of both pattern density and thickness on the curvature is less pronounced in the negative range of H_p . Another interesting observation from Figs 5.2 and 5.3 in the negative range of H_p is that the dependence of the curvature κ_{11} on H_p can be non-monotonic, i.e., as H_p decreases from zero, the curvature would increase initially but could start decreasing as H_p gets more negative. This nonmonotonic response can be attributed to the competition between strain energy and thickness reduction. The continuous decrease (i.e., more negative) in H_p leads to effective reduction in the overall nanomembrane thickness which facilitates rolling and favors an increase in the curvature, while at the same time it relieves the strain energy, contributing to lower mechanical driving force for the rolling. Therefore, the local maximum the rolling curvature indicates the point where the benefit from thickness reduction is offset by the loss in strain energy density.

Material	Young's modulus <i>E</i> (GPa)	Poisson's ratio (v)	Thicknesses $(h_t \text{ or } h_b) \text{ (nm)}$	Mismatch strain (ε_0)
GaAs	85.6	0.3	5.2	1 / 20/
In _{0.2} Ga _{0.8} As	75.1	0.3	5.8	1.4370
GaN	252	0.26	10	2 / 1%
AlN	297	0.3	10	2.4170

Table 5.1. The relevant material properties of the two nanomembrane systems, $GaAs^{(top)}/In_{0.2}Ga_{0.8}As^{(bottom)}$ and $GaN^{(top)}/AlN^{(bottom)}$ [22, 53, 56] considered in the present study.



Figure 5.2. Comparison between the FE simulated (symbols) and predicted (lines, from Eqs. (5.6-9)) rolling curvature κ_{11} as the pattern thickness H_p and pattern densities (ρ_1 , ρ_2) vary, for the GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom) nanomembrane system with (a) grating and (b) rectangular patterns.



Figure 5.3. Comparison between the FE simulated (symbols) and predicted (lines, from Eqs. (5.6-9)) rolling curvature κ_{11} as the pattern thickness H_p and pattern densities (ρ_1 , ρ_2) vary, for the GaN^(top)/AlN^(bottom) nanomembrane system with (a) grating and (b) rectangular patterns.

5.4.2 Flexible tuning of rolling direction and roll-up geometry via patterning

Recent experiments on grating-patterned metallic micro-springs reported that the rolling direction of the patterned grating structures in general stays consistently perpendicular to the long side edge of the grating pattern [25], in strong contrast to the rolling behaviors of nanomembranes without surface modifications, which typically exhibit long-edge rolling if not constrained [57, 58]. This indicates the strong ability of grated patterns in regulating the rolling direction of nanomembranes. Nonetheless, so far there exists no dedicated theoretical work to study such regulation. In this regard, here we performed FE simulations, considering a

rectangular nanomembrane with grating patterns arranged along the lateral direction (i.e., $\rho_1 > 0$, $\rho_2 = \infty$), as illustrated in Fig. 5.4. It was found that the grating patterns, of sufficient thickness (H_p) and density (ρ_1), can transform the originally long-edge (lateral) rolling mode (cf. Figs 5.4(a)) into short-edge (longitude) rolling mode (cf. Fig. 5.4(c)). More comprehensive FE investigation reveals that the resultant roll-up direction depends on not only the thickness and density of the grating patterns, but also the aspect (i.e., length/width) ratio R of nanomembranes, as demonstrated in Fig. 5.5(a). From Fig. 5.5(a), the threshold (H_p , ρ_1) curves corresponding to the lateral-to-longitude rolling mode transition can be identified for nanomembranes of different aspect ratios, and it is interesting to see that those curves effectively coincide into a single curve for large aspect ratios (i.e., R > 4). This consolidated curve basically gives the requirement of H_p and ρ_1 that can always ensure nanomembrane rolling perpendicular to the grating pattern.

Another interesting point to note from Fig. 5.5(a) is that as the pattern density ρ_1 increases, the H_p required to induce the rolling mode transition quickly decreases and stays nearly a constant value for large ρ_1 values. Given that the grating pattern serves to induce rolling anisotropy (i.e., promoting longitude rolling over lateral rolling), this indicates that the effect of patterning is more prominent at low pattern density. To further understand the above observation, we examine the roll-up curvatures κ_{11} and κ_{22} (predicted from Eqs (5.6-9)) for the longitude and lateral rolling respectively, and define a parameter $\Lambda = (\kappa_{22} - \kappa_{11})/\kappa_{11}$. The parameter Λ effectively characterizes the magnitude of anisotropy in rolling, and reflects on the different driving forces along the two rolling directions. Fig. 5.5(b) plots Λ as a function of ρ_1 for different H_p . We note that the Λ vs. ρ_1 curve gets elevated as H_p increases. For all H_p , Λ is seen to first rapidly increase with the pattern density, and then gradually decay with further increase in the density. The decline in Λ at large ρ_1 suggests that excess pattern density have little influence on rolling anisotropy. Comparing Figs 5.5(a) and 5.5(b), we can see that the

turning point (in the trend) in the Λ vs. ρ_1 curve roughly coincides with the location where the H_p vs. ρ_1 curve starts to level out.



Figure 5.4. FE simulated results of the rolling behaviours for sample rectangular nanomembranes (a) without and (b and c) with grating patterns, with patterns indicated by orange strips, showing the transition of the rolling model from the long-edge (lateral) rolling (a) without or (c) with insufficient grating patterning to short-edge (longitude) rolling (b) with sufficient presence of grating patterns. In this illustrative example, the nanomembrane is assumed to be of $GaAs^{(top)}/In_{0.2}Ga_{0.8}As^{(bottom)}$. Sample FE simulation movies illustrating the complete roll-up process available in Supplementary Material.



Figure 5.5. (a) Diagram of preferred rolling modes for the strained, grating-patterned nanomembranes of different aspect ratio *R* as the H_p and ρ_1 vary, where the regimes above and below the curve (for a particular *R*) correspond to short-edge (longitude) and long-edge (lateral) rolling respectively. (b) The magnitude of rolling anisotropy between lateral and longitudinal directions are quantitatively measured using $\Lambda = (k_{la} - k_{lo})/k_{lo}$ as a function of ρ and H_p . (c) shows a schematic illustration of two rolling modes (respectively corresponding to short-edge and long-edge rolling) with grating pattern structures. The patterned and normal strained bilayer are colored in gold, green and blue, respectively. In this illustrative example, the nanomembrane material system is assumed to be GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom).

The predictive mapping of rolling modes presented in Fig. 5.5 (a) provides critical guidance for the design of grating structures on nanomembranes, particularly in cases where the regulation of rolling directions is important. Moreover, these insights are also transferable to the design of more complex roll-up geometries. For instance, the predictive design of a helical configuration can be achieved from a grating-patterned rectangular nanomembrane template, as illustrated in Figs. 5.6 and 7 and elaborated in detail below.

For a grating-patterned rectangular nanomembrane, the rolling direction can be precisely controlled by choosing the appropriate H_p - ρ_1 combination (cf. Fig. 5.5). In particular,

for short-edge (longitude) rolling, the grating pattern stays perpendicular to the rolling direction. Now let's consider a strip cut from the nanomembrane, aligned at an angle α to the longitude direction (see Fig. 5. 6(a)). A series of FE simulations were performed to examine the formation of helical configurations from the rolling of such strips. It was found that the resultant helical configuration is strongly correlated with rolling behaviors of the host nanomembrane. In particular, the H_p - ρ_1 combination that ensures short-edge (longitude) rolling of the host nanomembranes also prescribes the rolling direction of the strip, leading to helix with axis parallel to the grating pattern, as illustrated in Fig. 5.7. Meanwhile, interestingly enough, these strips also exhibit nearly identical rolling diameter D with the host nanomembrane (and thus can be predicted by Eqs (5.6-9)), as demonstrated in Fig. 5.7 (b). With both the axis and diameter of the helix known, the pitch period (P) of the helix can also been determined:

$$P = \pi D \cdot \tan \alpha \tag{5.10}$$

where α denotes the helical angle, being also the angle that the strip makes with the longitude direction (cf. Fig. 5.6 (a)). On the other hand, within the H_{p} - ρ_1 domain where the host nanomembrane exhibits long-edge (lateral) rolling, the rolling of the cut strips is much more complex with great variation in the resultant rolling direction and diameter, showing no simple or apparent relation to the rolling of the host nanomembrane, and as a consequence, design of helixes with controlled configurations is not feasible (e.g., patterns show random alignment with respect to the helix axis and the helix shows non-straight axis and varying curvature spatially).

The above results demonstrate a new route to achieve predictive design of helixes with precise control of their geometry. The fact that we can use a pre-designed, grating patterned rectangular nanomembrane as the template also provides interesting design insights towards versatile and flexible fabrication of 3D roll-up nanostructures.



Figure 5.6. Schematic illustration of (a) strips cut from a grating-patterned host nanomembrane, aligned at different angles α (e.g., $\alpha = 37^{\circ}$, 45° and 57°) to the direction perpendicular to the grating pattern, which, after roll-up (with the rolling direction indicated by the black dashed arrows in (a)), results in (b) helixes with axis parallel to the grating pattern (obtained from FE simulations). (b) FE simulated helical strips with designed helical angles ($\alpha = 37^{\circ}, 45^{\circ}, 57^{\circ}$). (c) and (d) plot the designed helix diameter D_{pred} and pitch period P_{pred} (from analytical predictions) against the ones (i.e., denoted as D_{sim} and P_{sim} respectively) obtained from FE simulations, showing excellent agreement. In this illustrative example, the nanomembrane material system is assumed to be GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom). List of data in plots (c) and (d) available in Supplementary Material.



Figure 5.7. The predictive design diagram of helical configurations formed from the rolling of strips cut from strained patterned nanomembranes as the H_p and ρ_1 vary. The black solid line in the diagram indicates the threshold (H_p , ρ_1) curve corresponding to the lateral-to-longitude rolling mode transition (for $R \ge 4$, also see Fig. 5.5a). The blue circle and red x-mark symbols indicate whether or not the resultant helical configuration is a controlled one with the helix diameter and pitch period well predicted. In this illustrative example, the nanomembrane material system is assumed to be GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom). List of data available in Supplementary Material.

5.5 Conclusion

Employing both analytical modeling and comprehensive finite-element (FE) simulations, we systematically studied the effect of topological patterning (i.e., grating and rectangular patterns) on the self-rolling behaviors of heteroepitaxial strained nanomembranes. Based on Von-Karman nonlinear theory and Ritz method, an analytical modeling framework that is capable of predicting the resultant curvature of the patterned nanomembrane has been formulated, with the accuracy and reliability of model prediction validated through FE simulations. Particularly for grating-patterned nanomembranes, it was shown that the rolling direction can be precisely controlled by appropriate selection of the pattern thickness and density. Furthermore, the findings can also directly translate into valuable design insights for more complex roll-up geometries, such as helical structures. As an example, the predictive

design of helical structures of desired geometric metrics (i.e., helical angle, pitch period and diameter), resulted from strips cut from a host grating-patterned nanomembrane was demonstrated. The present study elucidates the pattern-induced potential anisotropic self-rolling behaviors and preferred rollup direction, providing new knowledge and mechanistic guidance towards predictive control and tuning of 3D roll-up nanostructures via topological patterning.

5.6 Acknowledgement

We greatly thank the financial support from McGill Engineering Doctoral Award, China Scholarship Council, National Sciences and Engineering Research Council (NSERC) Discovery grant (grant # RGPIN-2017-05187), and NSERC Strategic grant (grant # STPGP 494012-16). We also acknowledge Supercomputer Consortium Laval UQAM McGill and Eastern Quebec for providing computing power.

5.7 Supporting information

Table I (data for Fig. 5.6c): The relevant parameters for the strips used for examining the helical rolling (corresponding to the data points shown in Fig. 6c), where h_b and h_t respectively denote the thicknesses of the bottom and top layers of the host nanomembrane where the strips were cut, and ρ_1 denotes the density of the grating pattern, H_p represents dimensionless thickness parameter and α is the helical angle. D_{sim} and D_{pred} are the FE-simulated and theory predicted (from Eqs (6-9)) diameters of curvature of the helix resulted from the rolling of the strip. Note that the material system is assumed to be GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom), and the nanomembrane is assumed to be of a length/width ratio R = 4. The strips have a fixed width of 15 μ m (however it is important to note that the roll-up curvature is not sensitive to the strip width).

Pattern Density ρ_1	H_p	Helical angle	Thickness h_b (nm)	Thickness h_t (nm)	$D_{ m pred}$ (μ m)	D _{sim} (µm)
1	1	37°	15	15	2.72	2.69
1	1.25	45°	15	15	2.85	2.77
1	1	57°	15	15	2.72	2.84
1	1	37°	20	20	3.63	3.64
1	1	37°	25	25	4.53	4.44
1	1.25	37°	25	25	4.75	4.86
2	1	37°	15	15	2.99	2.97
2	1	57°	15	15	2.99	3.04
2	1	37°	20	20	3.98	3.90
2	1	37°	25	25	4.97	4.93
2	1	45°	25	25	4.97	4.83
3	1	37°	15	15	3.14	3.08
3	1	57°	15	15	3.14	3.07
3	1.25	37°	15	15	3.22	3.35
3	1	37°	20	20	4.18	4.24
3	1.25	37°	20	20	4.31	4.42
3	1	37°	25	25	5.23	5.17

Table II (data for Fig. 5.6 d): The relevant parameters for the strips used for examining the helical rolling (corresponding to the data points shown in Fig. 6d), where h_b and h_t respectively denote the thicknesses of the bottom and top layers of the host nanomembrane where the strips were cut, ρ_1 denotes the density of the grating pattern and H_p represents dimensionless thickness parameter and α is the helical angle. P_{sim} and P_{pred} are the FE-simulated and theory predicted (from Eq (6-10)) diameters of curvature of the helix resulted from the rolling of the strip. Note that the material system is assumed to be GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom), and the nanomembrane is assumed to be of a length/width ratio R = 4. The strips have a fixed width of 15 μ m (however it is important to note that the roll-up curvature is not sensitive to the strip width).

Pattern Density ρ_1	H _p	Helical angle	Thickness h_b (nm)	Thickness h_t (nm)	$P_{ m pred}$ (μ m)	P _{sim} (μm)
1	1	37°	15	15	6.44	6.37
1	1.25	45°	15	15	8.96	8.7
1	1	57°	15	15	13.16	13.72
1	1	37°	20	20	8.59	8.61
1	1	37°	25	25	10.74	10.51
1	1.25	37°	25	25	11.5	11.25
2	1	37°	15	15	7.07	7.03
2	1	57°	15	15	14.45	14.7
2	1	37°	20	20	9.43	9.23
2	1	37°	25	25	11.79	11.68
2	1	45°	25	25	15.64	15.17
3	1	37°	15	15	7.44	7.28
3	1	57°	15	15	15.2	14.85
3	1.25	37°	15	15	7.93	7.63
3	1	37°	20	20	9.91	10.03
3	1.25	37°	20	20	10.46	10.18
3	1	37°	25	25	12.39	12.25

Table III (data for Fig. 5.7): The relevant parameters for the strips used for examining the helical rolling (corresponding to the data points shown in Fig. 7), where h_b and h_t respectively denote the thicknesses of the bottom and top layers of the host nanomembrane where the strips were cut, ρ_1 denotes the density of the grating pattern and H_p represents dimensionless thickness parameter. Note that the material system is assumed to be GaAs^(top)/In_{0.2}Ga_{0.8}As^(bottom), and the nanomembrane is assumed to be of a length/width ratio R = 4. The strips have a fixed width of 15 μ m (however it is important to note that the roll-up curvature is not sensitive to the strip width).

Pattern Density ρ_1	Helical angle	Thickness h_b (nm)	Thickness h_t (nm)	H_p	Controlled Rolling? FE-simulated Predicted (Y or N)	
0.5	45°	25	25	1.80	Y	Y
0.5	37°	25	25	1.48	Y	Y
0.5	37°	25	25	1.28	Y	Y
0.5	37°	25	25	0.88	Y	Ν
0.5	37°	25	25	0.72	N	Ν
0.5	57°	25	25	0.56	N	Ν
1	57°	25	25	1.24	Y	Y
1	37°	25	25	1.08	Y	Y
1	37°	25	25	0.92	Y	Y
1	37°	25	25	0.76	Y	Y
1	37°	25	25	0.64	Ν	Ν
1	57°	25	25	0.48	N	Ν
1	45°	25	25	0.32	Ν	Ν
2	37°	25	25	1.04	Y	Y
2	45°	25	25	0.88	Y	Y
2	37°	25	25	0.72	Y	Y
2	37°	25	25	0.60	N	Ν
2	57°	25	25	0.44	N	Ν
2	37°	25	25	0.28	N	Ν
3	45°	25	25	1.04	Y	Y
3	37°	25	25	0.88	Y	Y
3	37°	25	25	0.72	Y	Y
3	37°	25	25	0.60	N	Ν
3	37°	25	25	0.48	N	Ν
3	57°	25	25	0.32	N	Ν
4	37°	25	25	0.98	Y	Y
4	37°	25	25	0.88	Y	Y
4	45°	25	25	0.72	Y	Ν
4	37°	25	25	0.56	Ν	Ν
4	45°	25	25	0.40	Ν	Ν
4	37°	25	25	0.24	N	N

5.8 References

[1] S. Janbaz, R. Hedayati, A.A. Zadpoor, Programming the Shape-Shifting of Flat Soft Matter: From Self-Rolling/Self-Twisting Materials to Self-Folding Origami, Materials Horizons, 3 (2016) 536-547.

[2] S. Sanchez, Lab-in-a-Tube Systems as Ultra-Compact Devices, Lab Chip, 15 (2015) 610-613.

[3] E.J. Smith, W. Xi, D. Makarov, I. Monch, S. Harazim, V.A.B. Quinones, C.K. Schmidt, Y.F. Mei, S. Sanchez, O.G. Schmidt, Lab-in-a-Tube: Ultracompact Components for on-Chip Capture and Detection of Individual Micro-/Nanoorganisms, Lab Chip, 12 (2012) 1917-1931.
[4] O.G. Schmidt, K. Eberl, Nanotechnology - Thin Solid Films Roll up into Nanotubes, Nature, 410 (2001) 168-168.

[5] V.Y. Prinz, V.A. Seleznev, A.K. Gutakovsky, A.V. Chehovskiy, V.V. Preobrazhenskii, M.A. Putyato, T.A. Gavrilova, Free-Standing and Overgrown Ingaas/Gaas Nanotubes, Nanohelices and Their Arrays, Physica E, 6 (2000) 828-831.

[6] D.J. Bell, L.X. Dong, B.J. Nelson, M. Golling, L. Zhang, D. Grutzmacher, Fabrication and Characterization of Three-Dimensional Ingaas/Gaas Nanosprings, Nano Lett, 6 (2006) 725-729.
[7] T. Ma, H.S. Liang, G. Chen, B. Poon, H.Q. Jiang, H.B. Yu, Micro-Strain Sensing Using Wrinkled Stiff Thin Films on Soft Substrates as Tunable Optical Grating, Optics Express, 21 (2013) 11994-12001.

[8] P. Cendula, A. Malachias, C. Deneke, S. Kiravittaya, O.G. Schmidt, Experimental Realization of Coexisting States of Rolled-up and Wrinkled Nanomembranes by Strain and Etching Control, Nanoscale, 6 (2014) 14326-14335.

[9] J. Wang, T.R. Zhan, G.S. Huang, P.K. Chu, Y.F. Mei, Optical Microcavities with Tubular Geometry: Properties and Applications, Laser Photonics Rev, 8 (2014) 521-547.

[10] S.L. Li, L.B. Ma, H.L. Zhen, M.R. Jorgensen, S. Kiravittaya, O.G. Schmidt, Dynamic Axial Mode Tuning in a Rolled-up Optical Microcavity, Appl Phys Lett, 101 (2012).

[11] S.W. Tang, Y.F. Fang, Z.W. Liu, L. Zhou, Y.F. Mei, Tubular Optical Microcavities of Indefinite Medium for Sensitive Liquid Refractometers, Lab Chip, 16 (2016) 182-187.

[12] S.Z. Huang, L. Zhang, X.Y. Lu, L.F. Liu, L.X. Liu, X.L. Sun, Y. Yin, S. Oswald, Z.Y. Zou, F. Ding, O.G. Schmidt, Tunable Pseudocapacitance in 3d Tio2-Delta Nanomembranes Enabling Superior Lithium Storage Performance, Acs Nano, 11 (2017) 821-830.

[13] L. Zhang, J.W. Deng, L.F. Liu, W.P. Si, S. Oswald, L.X. Xi, M. Kundu, G.Z. Ma, T. Gemming, S. Baunack, F. Ding, C.L. Yan, O.G. Schmidt, Hierarchically Designed Siox/Sioy Bilayer Nanomembranes as Stable Anodes for Lithium Ion Batteries, Adv Mater, 26 (2014) 4527-4532.

[14] C.L. Yan, W. Xi, W.P. Si, J.W. Deng, O.G. Schmidt, Highly Conductive and Strain-Released Hybrid Multilayer Ge/Ti Nanomembranes with Enhanced Lithium-Ion-Storage Capability, Adv Mater, 25 (2013) 539-544.

[15] H. Wang, M. Pumera, Fabrication of Micro/Nanoscale Motors, Chem Rev, 115 (2015) 8704-8735.

[16] Y.F. Mei, A.A. Solovev, S. Sanchez, O.G. Schmidt, Rolled-up Nanotech on Polymers: From Basic Perception to Self-Propelled Catalytic Microengines, Chem Soc Rev, 40 (2011) 2109-2119.

[17] D.a. Grimm, Rolled-up Nanomembranes as Compact 3d Architectures for Field Effect Transistors and Fluidic Sensing Applications, Nano Lett, 13 (2013) 213-218.

[18] O.G. Schmidt, C. Deneke, S. Kiravittaya, R. Songmuang, H. Heidemeyer, Y. Nakamura, R. Zapf-Gottwick, C. Muller, N.Y. Jin-Phillipp, Self-Assembled Nanoholes, Lateral Quantum-Dot Molecules, and Rolled-up Nanotubes, Ieee J Sel Top Quant, 8 (2002) 1025-1034. [19] D.J. Bell, L. Dong, B.J. Nelson, M. Golling, Fabrication and Characterization of Three-Dimensional Ingaas Gaas Nanosprings, Nano, (2006).

[20] Z. Tian, L. Zhang, Y.F. Fang, B. Xu, S.W. Tang, N. Hu, Z.H. An, C. Zi, Y.F. Mei, Deterministic Self-Rolling of Ultrathin Nanocrystalline Diamond Nanomembranes for 3d Tubular/Helical Architecture, Adv Mater, 29 (2017).

[21] Y.F. Mei, G.S. Huang, A.A. Solovev, E.B. Urena, I. Moench, F. Ding, T. Reindl, R.K.Y. Fu, P.K. Chu, O.G. Schmidt, Versatile Approach for Integrative and Functionalized Tubes by Strain Engineering of Nanomembranes on Polymers, Adv Mater, 20 (2008) 4085-4590.

[22] I.S. Chun, V.B. Verma, V.C. Elarde, S.W. Kim, J.M. Zuo, J.J. Coleman, X. Li, Ingaas/Gaas 3d Architecture Formation by Strain-Induced Self-Rolling with Lithographically Defined Rectangular Stripe Arrays, J Cryst Growth, 310 (2008) 2353-2358.

[23] X.L. Li, Strain Induced Semiconductor Nanotubes: From Formation Process to Device Applications, J Phys D Appl Phys, 41 (2008).

[24] Y.V. Nastaushev, V.Y. Prinz, S.N. Svitasheva, A Technique for Fabricating Au/Ti Microand Nanotubes, Nanotechnology, 16 (2005) 908-912.

[25] T. Huang, Z.Q. Liu, G.S. Huang, R. Liu, Y.F. Mei, Grating-Structured Metallic Microsprings, Nanoscale, 6 (2014) 9428-9435.

[26] W. Huang, X. Yu, P. Froeter, R.M. Xu, P. Ferreira, X.L. Li, On-Chip Inductors with Self-Rolled-up Sinx Nanomembrane Tubes: A Novel Design Platform for Extreme Miniaturization, Nano Lett, 12 (2012) 6283-6288.

[27] X. Yu, W. Huang, M.Y. Li, T.M. Comberiate, S.B. Gong, J.E. Schutt-Aine, X.L. Li, Ultra-Small, High-Frequency, and Substrate-Immune Microtube Inductors Transformed from 2d to 3d, Sci Rep-Uk, 5 (2015).

[28] R. Arayanarakool, A.K. Meyer, L. Helbig, S. Sanchez, O.G. Schmidt, Tailoring Three-Dimensional Architectures by Rolled-up Nanotechnology for Mimicking Microvasculatures, Lab Chip, 15 (2015) 2981-2989.

[29] Y.Y. Hu, W.M. Huang, Elastic and Elastic-Plastic Analysis of Multilayer Thin Films: Closed-Form Solutions, J Appl Phys, 96 (2004) 4154-4160.

[30] G.P. Nikishkov, Curvature Estimation for Multilayer Hinged Structures with Initial Strains, J Appl Phys, 94 (2003) 5333-5336.

[31] Y.C. Tsui, T.W. Clyne, An Analytical Model for Predicting Residual Stresses in Progressively Deposited Coatings .1. Planar Geometry, Thin Solid Films, 306 (1997) 23-33.

[32] M. Grundmann, Nanoscroll Formation from Strained Layer Heterostructures, Appl Phys Lett, 83 (2003) 2444-2446.

[33] J. Zang, M.H. Huang, F. Liu, Mechanism for Nanotube Formation from Self-Bending Nanofilms Driven by Atomic-Scale Surface-Stress Imbalance, Phys Rev Lett, 98 (2007).

[34] C. Cheng, S. Pengfei, M. Fanchao, L. Xiao, L. Xinyu, S. Jun, Nanotechnology, (2017).

[35] F. Cavallo, M.G. Lagally, Semiconductors Turn Soft: Inorganic Nanomembranes, Soft Matter, 6 (2010) 439-455.

[36] F. Cavallo, R. Songmuang, O.G. Schmidt, Fabrication and Electrical Characterization of Si-Based Rolled-up Microtubes, Appl Phys Lett, 93 (2008).

[37] F. Cavallo, R. Songmuang, C. Ulrich, O.G. Schmidt, Rolling up Sige on Insulator, Appl Phys Lett, 90 (2007).

[38] T. Kipp, H. Welsch, C. Strelow, C. Heyn, D. Heitmann, Optical Modes in Semiconductor Microtube Ring Resonators, Phys Rev Lett, 96 (2006).

[39] C. Deneke, C. Muller, N.Y. Jin-Phillipp, O.G. Schmidt, Diameter Scalability of Rolled-up in(Ga)as/Gaas Nanotubes, Semicond Sci Tech, 17 (2002) 1278-1281.

[40] N.Y. Jin-Phillipp, J. Thomas, M. Kelsch, C. Deneke, R. Songmuang, O.G. Schmidt, Electron Microscopy Study on Structure of Rolled-up Semiconductor Nanotubes, Appl Phys Lett, 88 (2006).

[41] F. Li, Z.T. Mi, Optically Pumped Rolled-up Ingaas/Gaas Quantum Dot Microtube Lasers, Optics Express, 17 (2009) 19933-19939.

[42] Z.B. Tian, V. Veerasubramanian, P. Bianucci, S. Mukherjee, Z.T. Mi, A.G. Kirk, D.V. Plant, Single Rolled-up Ingaas/Gaas Quantum Dot Microtubes Integrated with Silicon-on-Insulator Waveguides, Optics Express, 19 (2011) 12164-12171.

[43] Z.B. Tian, F. Li, Z.T. Mi, D.V. Plant, Controlled Transfer of Single Rolled-up Ingaas-Gaas Quantum-Dot Microtube Ring Resonators Using Optical Fiber Abrupt Tapers, Ieee Photonic Tech L, 22 (2010) 311-313.

[44] L.B. Freund, Substrate Curvature Due to Thin Film Mismatch Strain in the Nonlinear Deformation Range, J Mech Phys Solids, 48 (2000) 1159-1174.

[45] S. Vidoli, Discrete Approximations of the Foppl-Von Karman Shell Model: From Coarse to More Refined Models, Int J Solids Struct, 50 (2013) 1241-1252.

[46] M.L. Dano, M.W. Hyer, Thermally-Induced Deformation Behavior of Unsymmetric Laminates, Int J Solids Struct, 35 (1998) 2101-2120.

[47] M.W. Hyer, Calculations of the Room-Temperature Shapes of Unsymmetric Laminates, J Compos Mater, 15 (1981) 296-310.

[48] A.F. Bower, Applied Mechanics of Solids, CRC Press, 2009.

[49] O.A. Bauchau, J.I. Craig, Kirchhoff Plate Theory, in: O.A. Bauchau, J.I. Craig (Eds.) Structural Analysis, Springer Netherlands, Dordrecht, 2009, pp. 819-914.

[50] C.G. Diaconu, P.M. Weaver, A.F. Arrieta, Dynamic Analysis of Bi-Stable Composite Plates, J Sound Vib, 322 (2009) 987-1004.

[51] M. Brunetti, A. Vincenti, S. Vidoli, A Class of Morphing Shell Structures Satisfying Clamped Boundary Conditions, Int J Solids Struct, 82 (2016) 47-55.

[52] E.J. Barbero, Finite Element Analysis of Composite Materials Using Abaqustm, Taylor & Francis, 2013.

[53] C. Deger, E. Born, H. Angerer, O. Ambacher, M. Stutzmann, J. Hornsteiner, E. Riha, G. Fischerauer, Sound Velocity of Alxga1-Xn Thin Films Obtained by Surface Acoustic-Wave Measurements, Appl Phys Lett, 72 (1998) 2400-2402.

[54] M.H.T. Dastjerdi, M. Djavid, S. Arafin, X. Liu, P. Bianucci, Z. Mi, P.J. Poole, Optically Pumped Rolled-up Inas/Ingaasp Quantum Dash Lasers at Room Temperature, Semicond Sci Tech, 28 (2013).

[55] Y.F. Mei, D.J. Thurmer, C. Deneke, S. Kiravittaya, Y.F. Chen, A. Dadgar, F. Bertram, B. Bastek, A. Krost, J. Christen, T. Reindl, M. Stoffel, E. Coric, O.G. Schmidt, Fabrication, Self-Assembly, and Properties of Ultrathin Aln/Gan Porous Crystalline Nanomembranes: Tubes, Spirals, and Curved Sheets, Acs Nano, 3 (2009) 1663-1668.

[56] J. Nord, K. Albe, P. Erhart, K. Nordlund, Modelling of Compound Semiconductors: Analytical Bond-Order Potential for Gallium, Nitrogen and Gallium Nitride, J Phys-Condens Mat, 15 (2003) 5649-5662.

[57] C. Chen, P.F. Song, F.C. Meng, X. Li, X.Y. Liu, J. Song, Quantitative Analysis and Predictive Engineering of Self-Rolling of Nanomembranes under Anisotropic Mismatch Strain, Nanotechnology, 28 (2017).

[58] S. Alben, B. Balakrisnan, E. Smela, Edge Effects Determine the Direction of Bilayer Bending, Nano Lett, 11 (2011) 2280-2285.

Chapter 6: Misfit dislocation induced strain relaxation effect on self-rolling of strain-engineered nanomembranes

As elaborated in previous chapters, the mismatch strain within the nanomembrane is central to the self-rolling process. However, the mismatch strain may be partially relaxed by misfit dislocations, which are commonly generated at the interface within bilayer or multilayer nanomembranes during the deposition process. Therefore, it is important to account for the effect of those dislocations. This chapter depicts a systematic study that combines atomistic simulations and continuum mechanics to investigate strain relaxation effects induced by misfit dislocation, and subsequent influence on the self-rolling of strain-engineered nanomembranes.

• This chapter is in preparation:

Misfit dislocation induced strain relaxation effect on self-rolling of strain-engineered nanomembranes, to be submitted.

Cheng Chen, Pengfei Song, Fanchao Meng, Pengfei Ou, Xinyu Liu, J Song*

6.1 Abstract

Combining atomistic simulations and continuum modeling, the effects of misfit dislocations on strain relaxation and subsequently self-rolling of strain-engineered nanomembranes have been investigated. Two representative material systems including (GaN/In_{0.5}Ga_{0.5}N) of wurtzite lattice, and II–VI materials (CdTe/CdTe_{0.5}So_{.5}) of zinc-blend lattice were considered. The atomistic characteristics of dislocation and the resulting lattice distorting were first determined by generalized-stacking-fault energy (GSFE) profile and disregistry function obtained through Peierls-Nabarro (PN) model. Those properties were then used to calculate the accurate mismatch strain of those nanomembranes with thepresence of dislocations, and as inputs into von-Karman shell theory to quantitatively evaluate the effects on self-rolling curvature and anisotropy. The theoretical results were further confirmed by atomistic simulations of different crystal geometries and dislocation configurations. Our results provide effective ways to predict and design the rollup configurations of strain-engineered semiconductor nanomembrane containing misfit dislocations, induced by a relatively large mismatch strain and increasing thickness.

6.2 Introduction

Offering unique mechanical flexibility and geometrical variabilities along with excellent machinability, the rolled-up nanotechnology involving nanomembranes that consist of two or more strained layers has emerged as the focus of interest for numerous advanced applications including drug delivery [1, 2], optical microcavity [3-6], optoelectronics [7-9], artificial biomimetic structures [10-17], micro/nanoscale motors [18-20], and on-chip energy storage integrated microdevices [21-23], among others. The essential functional principle for the rolled-up nanotechnology relies on engineered strain gradient embedded in bi- or multilayer nanomembranes, which drives nanomembranes to self-roll into tubular geometries with tunable diameters once detached and relaxed from their host substrates [24]. For semiconductor nanomembranes, engineering of the strain gradient therein can be achieved through adjusting the composition dependent lattice mismatch of the strained layers utilizing heteroepitaxial deposition methods involving, e.g., molecular beam epitaxy (MBE) and metalorganic chemical vapour deposition (MOCVD) [25-27]. Both methods not only can precisely manipulate the composition and thickness of nanomembranes at the monolayer scale to enable strain gradient tuning, but also are applicable to a wide range of nanomembrane materials, such as Group IV (e.g., Si/Ge) [28-30], III–V (e.g., InGaN/GaN) [6, 26, 27, 31-34] and II–VI (e.g., CdS/CdSe) materials [35]. In contrast to the existing capability and flexibility in engineer the strain, accurate determination of the mismatch strain, on the other hand, can be challenging, particularly in cases of large strains [36] and film thickness [32, 34, 37, 38], as the mismatch strain could be partially relaxed via the nucleation of misfit dislocations at the interface, Consequently the strain gradient cannot be well evaluated by considering only lattice mismatch. Moreover, in some cases, especially for the epitaxial growth of highly mismatched materials, periodic interfacial misfit dislocations are intentionally introduced to effectively alleviate the mismatch strain and reduce the density of vertically propagating threading dislocations that are highly detrimental to device performance [39, 40]. However, despite the importance in affecting the strain in the nanomembrane, there has been limited work to investigate the dislocation induced strain relaxation and its subsequent effect on the self-rolling behaviors of strain engineered nanomembranes. Consequently, this deficit prevents the precise prediction and manipulation of the geometry of rolled-up nanomembranes where dislocations exist.

In this paper, we present a systematic study, aiming to address the afore-mentioned deficit in the understanding of the role of misfit dislocations on self-rolling of strain-engineered nanomembranes, combining atomistic simulations and continuum modeling. Two generic bilayer material models, the GaN/In_{0.5}Ga_{0.5}N (III–V materials) of wurtzite lattice, and CdTe/CdTe_{0.5}S_{0.5} (II–VI materials) of zinc-blend lattice, were chosen as representatives of nanomembrane systems in the presence study. Stable misfit dislocation configurations at the interface were explicitly constructed and examined using molecular dynamics simulations. The effects of strain relaxation and strain distribution induced by misfit dislocations were quantitatively determined by the disregistry functions obtained through Peierls-Nabarro (PN) model with the generalized-stacking-fault energy (GSFE) profiles of those dislocations as inputs. The effects of misfit dislocations on self-rolling curvatures of strain-engineered nanomembranes were then further evaluated employing von-Karman shell theory combined with the disregistry functions, validated by atomistic simulations. Our study provides a new theoretical approach to quantitatively account for the misfit dislocations induced strain relaxation in predictive engineering of self-rolling of strained nanomembranes.

6.3 Computational Methodology

Atomistic simulations of dislocations effects on self-rolling are performed in the framework of classical molecular dynamics (MD) implemented in the LAMMPS package.[41] The Stillinger-Weber potentials [42] have been employed for defining the interatomic forces of

two strain engineered bilayer nanomembranes, namely, CdTe/CdTe_{0.5}S_{0.5} and GaN/In_{0.5}Ga_{0.5}N. Those interatomic potentials have been demonstrated to correctly predict key physical properties, including the crystal structure, lattice constant, elastic modulus, and defect characteristics for the ternary systems of Cd-S-Te [43] and In-Ga-N [44] under stoichiometric conditions. The potentials of Cd-S-Te and In-Ga-N systems have been previously applied in the studies of strain relaxation of misfit dislocation [45, 46], heteroepitaxial growth [47], quantum dots [45] and substitutional diffusion of heterostructures [44].

Based on above potentials, the lattice constants have been determined to be (5.835 Å and 6.124 Å) for CdTe and CdTe0.5S0.5, while (3.175 Å and 3.348 Å) for GaN and In0.5Ga0.5N. These values are in good agreement with data reported in previous studies [46, 48]. The typical misfit dislocations identified in the heterostructures of CdTe/CdTe0.5S0.5 and GaN/In0.5Ga0.5N have Burgers vectors $\vec{b_1} = 1/2[011]$ and $\vec{b_2} = 1/3[11\overline{2}0]$ for the (001)[011] slip system and $(1\overline{1}00)[11\overline{2}0]$ slip system, respectively. To obtain the GSFEs of those dislocations, two supercells are constructed, as presented in Fig. 6.1, with orientations of $x_1 = [011], x_2 = [01\overline{1}], x_3 = [01\overline{1}], x_4 = [01\overline{1}], x_5 = [01\overline{1}], x_5$ and $x_3 = [001]$ for CdTe/CdTe_xS_{1-x}, while $x_1 = [11\overline{2}0]$, $x_2 = [0001]$, and $x_3 = [1\overline{1}00]$ for GaN/In_xGa_{1-x}N. The mismatch strain on the interface of CdS/CdTe_{0.5}S_{0.5} and GaN/In_{0.5}Ga_{0.5}N are then calculated to be 4.86% and 5.18%, respectively. The dimensions of supercells for CdTe/CdTe_{0.5}S_{0.5} and GaN/In_{0.5}Ga_{0.5}N are set to be 9.16 nm \times 9.16nm \times 12.96 nm, and 6.35 nm × 5.18nm × 7.70 nm along x_1 , x_2 and x_3 directions. To calculate the GSFEs, we adopt the relaxation procedures outlined in Refs. [49, 50]. First, the upper half crystal over the heterogeneous interface of nanomembranes (shaded in Fig. 6.1) are incrementally shifted with respect to the below part along the $-x_1$ direction, where atoms are fixed along the x_1 and x_2 directions but free to move along the x_3 direction. The attendant energy cost per unit area, i.e., the GSFE (denoted as γ_{GSF} below), is monitored as a function of the applied shift vector.



Figure 6.1. Three dimensional schematic illustrations of the (a) zinc blend CdS/CdTe_{0.5}S_{0.5} and (c) wurtzite GaN/In_{0.5}Ga_{0.5}N where the shaded plane indicated the interface. (b) and (d) Projection views of atomic configurations along [011] and [$11\overline{2}0$] to illustrate the local atom arrangements on the interface. Cd, Te and S atoms are colored in blue, red and yellow, respectively. The N atoms are indicated by small blue spheres, while In and Ga atoms are represented by purple and golden spheres.



Figure 6.2. (a) the 5&7-ring core configuration for the 1/2 [011](001) edge dislocation on the mismatched interface of CdS/CdTe_{0.5}S_{0.5} (projection view along $[01\overline{1}]$ direction) (b) the 8-ring core for the $1/3 [11\overline{2}0]$ edge dislocation residing in the mismatched interface of

GaN/In_{0.5}Ga_{0.5}N (projection view along [**0001**] direction). The atoms are colored similar to Fig. 6.1.

Besides simulating GSFEs, a separate set of large scale atomistic simulations were performed to compute the self-rolling curvature of strain-engineered nanomembranes with different misfit dislocations densities located on their interfaces. In those simulations, larger simulation boxes with varied dislocation density have been utilized with dimensions $L_y = 3300$ Å while L_x and L_z ranging from 62 Å to 366 Å, and 65 Å to 87 Å, respectively, containing ~3 million atoms. The misfit dislocations are first generated by displacing all atoms according to the isotropic linear elasticity theory, as shown in Fig. 6. 2. Two stable core structures of the misfit dislocations have been identified, being 5-7 atom ring core for the zinc-blend structure of CdTe/CdTe0.5S0.5, and 8 atom ring core for the wurtzite structure of GaN/In0.5Ga0.5N. Subsequently, the mismatched nanomembranes with those dislocation structures constructed are relaxed to be bending out of plane using the conjugate gradient algorithm [51]. The typical examples of rolling up structures after relaxation are presented in the Fig. 6.3, where the curvature radius is estimated by tracking the coordinates of atoms. The dislocation detections, visualization and analysis are conducted using the Ovito software [52]. In our simulations, the dislocation density is defined as the dislocation number per unit length along Burger's vector direction on the slip plane of dislocation. Therefore, the dislocation effects could be examined by controlling the distance between two closed dislocations.



Figure 6.3. The representative rollup structures of MD simulations with misfit dislocations distributed in the interface of mismatched nanomembranes. (a) and (b) indicate the self-rolled-up structures of CdS/CdTe_{0.5}S_{0.5} and GaN/In_{0.5}Ga_{0.5}N, respectively, while (c) shows a schematic illustration of (b) and highlights the corresponding dislocation lines using the dislocation detection algorithm in the Ovito software [52].

6.4 **Results and Discussion**

6.4.1 Effects of misfit dislocation on strain gradient of nanomembranes

The effects of misfit dislocation on strain gradient of nanomembranes are examined in detail using the disregistry functions of dislocation, describing the relative displacement between two half crystals divided by the heterogeneous interface of nanomembranes. The disregistry functions of dislocation are obtained through Peierls-Nabarro (PN) model [53, 54]

together with an input from the GSFEs of dislocations, calculated by atomistic simulations. Fig. 6.4 gives the GSFE curves of dislocations for the (001)[011] slip system of CdTe/CdTe_{0.5}S_{0.5} and (1100)[1120] slip system of GaN/In_{0.5}Ga_{0.5}N, where δ and *b* denote the shifting displacement and the magnitude of the Burgers vector. It is revealed that for all cases the GSFE curve exhibits a maximum, i.e., the unstable stacking fault at $\delta = b/2$, indicating that those misfit dislocations only have planar cores and cannot dissociate into partial dislocations. Afterwards, based on those GSFE curves, the disregistry function u(x) can be deduced by considering the balance between the stress induced by the dislocation and the periodic lattice restoring stress $-\frac{\partial \gamma_{GSFE}(u(x))}{\partial u(x)}$ that represents the lattice resistance of a crystal to the distortion

associated with the dislocation core:

$$K\int_{-\infty}^{\infty} \frac{\left(du(x)/dx\right)_{x=x'}dx'}{x-x'} = -\frac{\partial\gamma_{GSFE}\left(u(x)\right)}{\partial u(x)},\tag{6.1}$$

Where K is a constant, being $\mu/(4\pi)$ for a screw dislocation and $\mu/[4\pi(1-v)]$ for an edge dislocation, μ and v are the shear modulus along the slip plane of dislocation and Poisson's ratio, respectively. Because the misfit dislocations are located at the interface of nanomembranes, μ and v are calculated to be the average value of the system, namely, 38 GPa and 0.33 for CdTe/CdTe_{0.5}S_{0.5}, while 108 GPa and 0.25 for GaN/In_{0.5}Ga_{0.5}N. The disregistry functions of

dislocations u(x), and the associated disregistry density $\rho(x) = \frac{du(x)}{dx}$ are finally obtained,

based on the method of trial fitting functions, as depicted in Fig. 6.5. This implies that the distortion induced by the misfit dislocation could be expressed as an nonlinear continuous function of the coordinate x along the Burger's vector direction, and reaches the maximum value at the core region of dislocations. Therefore, the inelastic strain ε^{in} of the system that includes the mismatch strain at the heterogeneous interface of nanomembranes and distortion of misfit dislocations could be further determined through principle of superposition, as shown

in Eq, (6.2),

$$\varepsilon^{in} = \varepsilon_0 + \sum_{i=1}^n \frac{\partial u(x+(i-1)\cdot d^m)}{\partial x}$$
(6.2)

Here, ε_0 and *n* indicate original mismatch strain and the number of misfit dislocations, respectively. d^m represents the distance between two closed dislocations.



Figure 6.4. The GSFE curves for the (001)[011] slip system of CdTe/CdTe_{0.5}S_{0.5} and $(1\overline{1}00)[11\overline{2}0]$ slip system of GaN/In_{0.5}Ga_{0.5}N, respectively.



Figure 6.5. The disregistry u(x) and associated disregistry density $\rho(x)$ profiles for the (**001**)[**011**] slip system of CdTe/CdTe_{0.5}S_{0.5} and (**1100**)[**1120**] slip system of GaN/In_{0.5}Ga_{0.5}N, respectively.

6.4.2 Effects of misfit dislocation on the rollup curvature of nanomembranes

The study of dislocation effect on the rollup curvature of nanomembranes is based on a combined numerical methodology using von-Karman shell theory [55] and disregistry functions of dislocations, and Fig. 6.6 depicts a typical precursor geometry of strained

nanomembranes with misfit dislocations embedded to produce the roll-up structure.



Figure 6.6. Illustration for the geometrical sizes of nanomembranes with misfit dislocations embedded on the interface of strained bilayers. *L* and *W* define the longitudinal and transverse length along the x_1 and x_2 direction respectively.

Assuming a plane-stress formulation, the total potential energy of this system with misfit dislocations incorporated H, can be expressed as a function of the mechanical and geometrical properties of nanomembranes by:

$$H = \int_{0}^{L} \int_{0}^{W} \left(\int_{-\frac{h}{2}}^{\frac{h}{2}-h_{t}} Q_{ij}^{b} \varepsilon_{i}^{b} \varepsilon_{j}^{b} dx_{3} + \int_{\frac{h}{2}-h_{t}}^{\frac{h}{2}} Q_{ij}^{t} \varepsilon_{i}^{t} \varepsilon_{j}^{t} dx_{3} \right) dx_{2} dx_{1}$$
(6.3)

where *L*, *W*, and *h* are the length of the nanomembranes in the x_1 and x_2 directions, and its total thickness. (h_t, h_b) and (Q_{ij}^t, Q_{ij}^b) denote the thicknesses and transformed reduced elastic stiffnesses of top and bottom layers, respectively. $(\varepsilon_i^b, \varepsilon_j^b)$ and $(\varepsilon_i^t, \varepsilon_j^t)$ define the in-plane elastic strains in the bottom and top layer with *i*, *j* = 1, 2, 6 as per convention, given by

$$\varepsilon_1^b = \varepsilon_1^m + x_3 \kappa_1, \ \varepsilon_2^b = \varepsilon_2^m + x_3 \kappa_2 \tag{6.4}$$

$$\varepsilon_1^t = \varepsilon_1^m + x_3 \kappa_1 - \varepsilon_1^{in}, \ \varepsilon_2^t = \varepsilon_2^m + x_3 \kappa_2 - \varepsilon_2^{in} \tag{6.5}$$

 $(\varepsilon_1^{in}, \varepsilon_2^{in})$ represent the component of inelastic strains in the top layer, namely, the mismatch strain and the strain relieved by the misfit dislocation. The quantities ε_1^m , ε_2^m and κ_1 , κ_2 are the midplane strains and curvatures, respectively, defined by

$$\varepsilon_1^m = \frac{\partial u_1}{\partial x_1} + \frac{1}{2} \left(\frac{\partial u_3}{\partial x_1} \right)^2, \quad \varepsilon_2^m = \frac{\partial u_2}{\partial x_2} + \frac{1}{2} \left(\frac{\partial u_3}{\partial x_1} \right)^2 \tag{6.6}$$

$$\kappa_1 = \frac{\partial^2 u_3}{\partial x_1^2}, \ \kappa_2 = \frac{\partial^2 u_3}{\partial x_2^2} \tag{6.7}$$

where u_i (*i*=1, 2, 3) are the midplane displacements of the nanomembranes in the x_1 , x_2 and x_3 directions. Here the extensional midplane strains are approximated using the following set of complete polynomials:

$$\varepsilon_1^m = \sum_{i,j=0}^n c_{ij} x_1^i x_2^j, \ \varepsilon_2^m = \sum_{i,j=0}^n d_{ij} x_1^i x_2^j \tag{6.8}$$

where the c_{ij} and d_{ij} are to-be-determined coefficients. In our calculations, an approximated displacement field is assumed for u_3 based on the Ritz method [56-58] as

$$u_3 = \frac{1}{2}(ax_1^2 + bx_2^2), \tag{6.9}$$

Where, *a* and *b* are parameters to be determined. Then, the in-plane shear strain $(\varepsilon_6^b, \varepsilon_6^b)$ could be further derived by strain--displacement relations. The coefficients and curvatures can be obtained by minimizing the potential energy *H*. According to the lattice constants and elastic modulus provided by MD simulations, the transformed reduced elastic stiffnesses of nanomembranes used in the numerical calculations are derived and presented in the table 6. 1.

Elastic Stiffness (GPa)	<i>Q</i> ₁₁	Q ₂₂	<i>Q</i> ₁₂	Q_{66}
GaN	328	351	69	115
Ino.5Ga0.5N	255	275	60	85
CdTe	72	72	19	30
CdTe0.5S0.5	47	47	16	24

Table 6.1. The reduced elastic stiffnesses obtained for CdTe/CdTe0.5S0.5 and GaN/In0.5Ga0.5N

We then start to examine the roll-up curvatures of nanomembranes with dislocations incorporated, and to determine the preferred rolling direction. As examples, we calculate the equilibrium strain energies of CdTe/CdTe_{0.5}S_{0.5} and GaN/In_{0.5}Ga_{0.5}N systems for both rolling up states along $-x_1$ and $-x_2$ directions, depending on the misfit dislocation densities, as given in Fig. 6.7.



Figure 6.7. Strain energies of the rollup structure of CdTe/CdTe_{0.5}S_{0.5} and GaN/In_{0.5}Ga_{0.5}N systems normalized on the strain energy of the flat structure are plotted against misfit dislocation densities for rolling up along $-x_1$ and $-x_2$ directions.

Obviously, the strain energy remains smallest for the $-x_1$ rolling directions for both cases, thus the $-x_1$ direction would be the preferred rolling direction. This can be explained by the strain relaxation along $-x_2$ direction (the Burger's vector direction) induced by misfit dislocations, resulting in the reduced bend force along this direction. Therefore, only the rolling curvatures κ_1 as the function of misfit dislocation density and thickness are calculated and compared with results obtained by MD simulations, as illustrated in Fig. 6.8. Here, the calculated curvatures are all converted into the rolling diameter that could be easy to be compared with those values obtained by tracking and measured the atomic coordinates in our MD simulations. In Fig. 6.8, we see that for all cases the increased dislocation density would lead to an overall increase of rolling diameters, which could explain the discrepancy between experimental measurements and traditional theoretical prediction using Timoshenko formula. In addition, the MD simulated results presented confirm the predictions obtained from the analytical models and the analytical model is reliable when applicable into the nanomembranes with different crystal symmetries and misfit dislocation configurations.



Figure 6.8. The predicted and MD simulated rolling diameter as a function of the misfit dislocation density for (a) CdTe/CdTe_{0.5}S_{0.5} and (b) GaN/In_{0.5}Ga_{0.5}N, respectively.

6.5 Conclusion

This work presents a combined framework that connects the atomic length scale variations of dislocations to continuum elasticity, and quantitatively evaluates the dislocations effects on the self-rolling of strain-engineered nanomembranes. The typical misfit dislocations
of (GaN/In0.5Ga0.5N) with wurtzite lattice, and (CdTe/CdTe0.5S0.5) with zinc-blend lattice have been created in MD simulations and their generalized stacking fault energy (GSFE) curves are calculated to be an input of PN model to obtain the disregistry functions of dislocations that accurately determine the lattice elastic distortion of dislocations. Afterwards, dislocation effects on the self-rolling behaviors of strain-engineered nanomembranes are further investigated employing von-Karman shell theory with the disregistry functions incorporated. It is found that, besides the elastic anisotropy of material, the formation of misfit dislocations could be another potential contributing factor causing the anisotropic rolling of strain-engineered nanomembranes. Additionally, we can see that independent of the thickness, the increased dislocation density would enhance the rollup diameters of released nanomembranes. The theoretical predictions are confirmed by the MD simulations of rolling up of nanomembranes with the existence of misfit dislocations, and the analytical method is shown to be reliable when applicable into different crystal geometries and dislocation configurations. Our work helps precise design the rollup structures from pre-defined geometries of strain-engineered nanomembranes with the consideration of dislocations especially for the situations of large mismatched strain and increasing thickness, and reduce the deviations of theoretical prediction for experimental observations.

6.6 Acknowledgement

We greatly thank the financial support from McGill Engineering Doctoral Award, China Scholarship Council, National Sciences and Engineering Research Council (NSERC) Discovery grant (grant # RGPIN-2017-05187), and NSERC Strategic grant (grant # STPGP 494012-16). We also acknowledge Supercomputer Consortium Laval UQAM McGill and Eastern Quebec for providing computing power.

6.7 References

[1] R. Arayanarakool, A.K. Meyer, L. Helbig, S. Sanchez, O.G. Schmidt, Tailoring Three-Dimensional Architectures by Rolled-up Nanotechnology for Mimicking Microvasculatures, Lab Chip, 15 (2015) 2981-2989.

[2] R. Fernandes, D.H. Gracias, Self-Folding Polymeric Containers for Encapsulation and Delivery of Drugs, Adv Drug Deliver Rev, 64 (2012) 1579-1589.

[3] G.S. Huang, Y.F. Mei, Electromagnetic Wave Propagation in a Rolled-up Tubular Microcavity, J Mater Chem C, 5 (2017) 2758-2770.

[4] J. Wang, T.R. Zhan, G.S. Huang, P.K. Chu, Y.F. Mei, Optical Microcavities with Tubular Geometry: Properties and Applications, Laser Photonics Rev, 8 (2014) 521-547.

[5] X.L. Li, Self-Rolled-up Microtube Ring Resonators: A Review of Geometrical and Resonant Properties, Adv Opt Photonics, 3 (2011) 366-387.

[6] T. Kipp, H. Welsch, C. Strelow, C. Heyn, D. Heitmann, Optical Modes in Semiconductor Microtube Ring Resonators, Phys Rev Lett, 96 (2006).

[7] S.M. Harazim, V.A.B. Quinones, S. Kiravittaya, S. Sanchez, O.G. Schmidt, Lab-in-a-Tube: On-Chip Integration of Glass Optofluidic Ring Resonators for Label-Free Sensing Applications, Lab Chip, 12 (2012) 2649-2655.

[8] a. Bernardi, S. Kiravittaya, a. Rastelli, R. Songmuang, D.J. Thurmer, M. Benyoucef, O.G. Schmidt, On-Chip Si/Siox Microtube Refractometer, Appl Phys Lett, 93 (2008).

[9] S.M. Harazim, W. Xi, C.K. Schmidt, S. Sanchez, O.G. Schmidt, Fabrication and Applications of Large Arrays of Multifunctional Rolled-up Sio/Sio2 Microtubes, Journal of Materials Chemistry, 22 (2012) 2878.

[10] K.U. Jeong, J.H. Jang, D.Y. Kim, C. Nah, J.H. Lee, M.H. Lee, H.J. Sun, C.L. Wang, S.Z.D. Cheng, E.L. Thomas, Three-Dimensional Actuators Transformed from the Programmed Two-Dimensional Structures Via Bending, Twisting and Folding Mechanisms, Journal of Materials Chemistry, 21 (2011) 6824-6830.

[11] L. Ionov, Biomimetic Hydrogel-Based Actuating Systems, Advanced Functional Materials, 23 (2013) 4555-4570.

[12] L. Persano, A. Camposeo, D. Pisignano, Integrated Bottom-up and Top-Down Soft Lithographies and Microfabrication Approaches to Multifunctional Polymers, J Mater Chem C, 1 (2013) 7663-7680.

[13] R. Kempaiah, Z.H. Nie, From Nature to Synthetic Systems: Shape Transformation in Soft Materials, J Mater Chem B, 2 (2014) 2357-2368.

[14] M. Podgorski, D.P. Nair, S. Chatani, G. Berg, C.N. Bowman, Programmable Mechanically Assisted Geometric Deformations of Glassy Two-Stage Reactive Polymeric Materials, Acs Appl Mater Inter, 6 (2014) 6111-6119.

[15] A.R. Studart, Biologically Inspired Dynamic Material Systems, Angew Chem Int Edit, 54 (2015) 3400-3416.

[16] F.M. Wisser, B. Schumm, G. Mondin, J. Grothe, S. Kaskel, Precursor Strategies for Metallic Nano- and Micropatterns Using Soft Lithography, J Mater Chem C, 3 (2015) 2717-2731.

[17] A.S. Gladman, E.A. Matsumoto, R.G. Nuzzo, L. Mahadevan, J.A. Lewis, Biomimetic 4d Printing, Nat Mater, 15 (2016) 413-418.

[18] H. Wang, M. Pumera, Fabrication of Micro/Nanoscale Motors, Chem Rev, 115 (2015) 8704-8735.

[19] J.X. Li, J. Zhang, W. Gao, G.S. Huang, Z.F. Di, R. Liu, J. Wang, Y.F. Mei, Dry-Released Nanotubes and Nanoengines by Particle-Assisted Rolling, Adv Mater, 25 (2013) 3715-3721.

[20] A.A. Solovev, Y.F. Mei, E.B. Urena, G.S. Huang, O.G. Schmidt, Catalytic Microtubular Jet Engines Self-Propelled by Accumulated Gas Bubbles, Small, 5 (2009) 1688-1692.

[21] X.F. Wang, Y. Chen, O.G. Schmidt, C.L. Yan, Engineered Nanomembranes for Smart Energy Storage Devices, Chem Soc Rev, 45 (2016) 1308-1330.

[22] X.H. Liu, J. Zhang, W.P. Si, L.X. Xi, B. Eichler, C.L. Yan, O.G. Schmidt, Sandwich Nano Architecture of Si/Reduced Graphene Oxide Bilayer Nanomembranes for Li-Ion Batteries with Long Cycle Life, Acs Nano, 9 (2015) 1198-1205.

[23] L. Zhang, J.W. Deng, L.F. Liu, W.P. Si, S. Oswald, L.X. Xi, M. Kundu, G.Z. Ma, T. Gemming, S. Baunack, F. Ding, C.L. Yan, O.G. Schmidt, Hierarchically Designed Siox/Sioy Bilayer Nanomembranes as Stable Anodes for Lithium Ion Batteries, Adv Mater, 26 (2014) 4527-4532.

[24] Z. Chen, G.S. Huang, I. Trase, X.M. Han, Y.F. Mei, Mechanical Self-Assembly of a Strain-Engineered Flexible Layer: Wrinkling, Rolling, and Twisting, Phys Rev Appl, 5 (2016).

[25] I.S. Chun, A. Challa, B. Derickson, K.J. Hsia, X.L. Li, Geometry Effect on the Strain-Induced Self-Rolling of Semiconductor Membranes, Nano Lett, 10 (2010) 3927-3932.

[26] I.S. Chun, V.B. Verma, V.C. Elarde, S.W. Kim, J.M. Zuo, J.J. Coleman, X. Li, Ingaas/Gaas 3d Architecture Formation by Strain-Induced Self-Rolling with Lithographically Defined Rectangular Stripe Arrays, J Cryst Growth, 310 (2008) 2353-2358.

[27] C. Deneke, C. Muller, N.Y. Jin-Phillipp, O.G. Schmidt, Diameter Scalability of Rolled-up in(Ga)as/Gaas Nanotubes, Semicond Sci Tech, 17 (2002) 1278-1281.

[28] F. Cavallo, M.G. Lagally, Semiconductors Turn Soft: Inorganic Nanomembranes, Soft Matter, 6 (2010) 439-455.

[29] F. Cavallo, R. Songmuang, O.G. Schmidt, Fabrication and Electrical Characterization of Si-Based Rolled-up Microtubes, Appl Phys Lett, 93 (2008).

[30] F. Cavallo, R. Songmuang, C. Ulrich, O.G. Schmidt, Rolling up Sige on Insulator, Appl Phys Lett, 90 (2007).

[31] N.Y. Jin-Phillipp, J. Thomas, M. Kelsch, C. Deneke, R. Songmuang, O.G. Schmidt, Electron Microscopy Study on Structure of Rolled-up Semiconductor Nanotubes, Appl Phys Lett, 88 (2006).

[32] F. Li, Z.T. Mi, Optically Pumped Rolled-up Ingaas/Gaas Quantum Dot Microtube Lasers, Optics Express, 17 (2009) 19933-19939.

[33] Z.B. Tian, V. Veerasubramanian, P. Bianucci, S. Mukherjee, Z.T. Mi, A.G. Kirk, D.V. Plant, Single Rolled-up Ingaas/Gaas Quantum Dot Microtubes Integrated with Silicon-on-Insulator Waveguides, Optics Express, 19 (2011) 12164-12171.

[34] Z.B. Tian, F. Li, Z.T. Mi, D.V. Plant, Controlled Transfer of Single Rolled-up Ingaas-Gaas Quantum-Dot Microtube Ring Resonators Using Optical Fiber Abrupt Tapers, Ieee Photonic Tech L, 22 (2010) 311-313.

[35] J.Q. Hu, Y. Bando, J.H. Zhan, M.Y. Liao, D. Golberg, X.L. Yuan, T. Sekiguchi, Single-Crystalline Nanotubes of Iib-Vi Semiconductors, Appl Phys Lett, 87 (2005).

[36] Y.F. Mei, D.J. Thurmer, C. Deneke, S. Kiravittaya, Y.F. Chen, A. Dadgar, F. Bertram, B. Bastek, A. Krost, J. Christen, T. Reindl, M. Stoffel, E. Coric, O.G. Schmidt, Fabrication, Self-Assembly, and Properties of Ultrathin Aln/Gan Porous Crystalline Nanomembranes: Tubes, Spirals, and Curved Sheets, Acs Nano, 3 (2009) 1663-1668.

[37] X. Li, W. Huang, Rolled-up Transformer Structure for a Radiofrequency Integrated Circuit (Rfic), in, Google Patents, 2014.

[38] J. Wang, E.M. Song, C.L. Yang, L.R. Zheng, Y.F. Mei, Fabrication and Whispering Gallery Resonance of Self-Rolled up Gallium Nitride Microcavities, Thin Solid Films, 627 (2017) 77-81.

[39] S.H. Huang, G. Balakrishnan, M. Mehta, A. Khoshakhlagh, L.R. Dawson, D.L. Huffaker, P. Li, Epitaxial Growth and Formation of Interfacial Misfit Array for Tensile Gaas on Gasb, Appl Phys Lett, 90 (2007).

[40] S.H. Huang, G. Balakrishnan, A. Khoshakhlagh, A. Jallipalli, L.R. Dawson, D.L. Huffaker, Strain Relief by Periodic Misfit Arrays for Low Defect Density Gasb on Gaas, Appl Phys Lett, 88 (2006).

[41] S. Plimpton, Fast Parallel Algorithms for Short-Range Molecular-Dynamics, J Comput Phys, 117 (1995) 1-19.

[42] F.H. Stillinger, T.A. Weber, Computer-Simulation of Local Order in Condensed Phases of Silicon, Phys Rev B, 31 (1985) 5262-5271.

[43] X.W. Zhou, D.K. Ward, J.E. Martin, F.B. van Swol, J.L. Cruz-Campa, D. Zubia, Stillinger-Weber Potential for the Ii-Vi Elements Zn-Cd-Hg-S-Se-Te, Phys Rev B, 88 (2013).

[44] X.W. Zhou, R.E. Jones, J. Gruber, Molecular Dynamics Simulations of Substitutional Diffusion, Comp Mater Sci, 128 (2017) 331-336.

[45] X.W. Zhou, M.E. Foster, F.B. van Swol, J.E. Martin, B.M. Wong, Analytical Bond-Order Potential for the Cd-Te-Se Ternary System, J Phys Chem C, 118 (2014) 20661-20679.

[46] J. Gruber, X.W. Zhou, R.E. Jones, S.R. Lee, G.J. Tucker, Molecular Dynamics Studies of Defect Formation During Heteroepitaxial Growth of Ingan Alloys on (0001) Gan Surfaces, J Appl Phys, 121 (2017).

[47] X.W. Zhou, J.J. Chavez, S. Almeida, D. Zubia, Understanding Misfit Strain Releasing Mechanisms Via Molecular Dynamics Simulations of Cdte Growth on {112}Zinc-Blende Cds, J Appl Phys, 120 (2016).

[48] X.W. Zhou, D.K. Ward, J.A. Zimmerman, J.L. Cruz-Campa, D. Zubia, J.E. Martin, F. van Swol, An Atomistically Validated Continuum Model for Strain Relaxation and Misfit Dislocation Formation, J Mech Phys Solids, 91 (2016) 265-277.

[49] C. Chen, F.C. Meng, J. Song, Effects of Mg and Al Doping on Dislocation Slips in Gan, J Appl Phys, 119 (2016).

[50] C. Chen, F.C. Meng, J. Song, Core Structures Analyses of (a Plus C)-Edge Dislocations in Wurtzite Gan through Atomistic Simulations and Peierls-Nabarro Model, J Appl Phys, 117 (2015).

[51] J. Nocedal, S.J. Wright, Conjugate Gradient Methods, Springer, 2006.

[52] A. Stukowski, Visualization and Analysis of Atomistic Simulation Data with Ovito-the Open Visualization Tool, Model Simul Mater Sc, 18 (2010).

[53] F. Nabarro, Dislocations in a Simple Cubic Lattice, Proceedings of the Physical Society, 59 (1947) 256.

[54] R. Peierls, The Size of a Dislocation, Proceedings of the Physical Society, 52 (1940) 34-37.

[55] S. Vidoli, Discrete Approximations of the Foppl-Von Karman Shell Model: From Coarse to More Refined Models, Int J Solids Struct, 50 (2013) 1241-1252.

[56] L.B. Freund, Substrate Curvature Due to Thin Film Mismatch Strain in the Nonlinear Deformation Range, J Mech Phys Solids, 48 (2000) 1159-1174.

[57] M.L. Dano, M.W. Hyer, Thermally-Induced Deformation Behavior of Unsymmetric Laminates, Int J Solids Struct, 35 (1998) 2101-2120.

[58] M.W. Hyer, Calculations of the Room-Temperature Shapes of Unsymmetric Laminates, J Compos Mater, 15 (1981) 296-310.

Chapter 7: Effects of material heterogeneity on self-rolling of strain engineered membranes

Besides the effects of misfit dislocations discussed in Chapter 5, material heterogeneity can also influence the internal strain gradient of membranes, which is often resulted from incorporating new structural units into strained membranes. Therefore, understanding of the effects of material heterogeneity on self-rolling of strain engineered membranes is key to achieving a higher level of manipulation over the rolled-up geometry, or integrating extra functionality modules that enhance the performance of existing structures and devices. In this chapter, an analytical model has been developed to evaluate the effects of distribution of heterogeneous embedding elements on the self-rolling behaviors of strained membranes, validated through molecular dynamic (MD) simulations and finite element (FE) analysis. The theoretical predictions could be used for precisely generating helical shapes with desired rolling curvatures, chirality, and helix angles, showing great potential in designing complex 3D structures based on strained membranes through incorporation of heterogeneous elements.

• This chapter is in preparation:

Effects of material heterogeneity on self-rolling of strain engineered membranes.

Cheng Chen, Pengfei Song, Fanchao Meng, Pengfei Ou, Xinyu Liu, J Song*

7.1 Abstract

During recent decades, due to the great potential of self-rolling geometry of strained membranes in the applications of biomedical devices and optoelectronics, significant research efforts have been focused on exploring and developing the advanced fabrication techniques that incorporate new structural units into membranes allowing tunable internal strain and achieving a higher level of manipulation over the rolled up geometry, or integrate extra functionality modules, thus enhancing the performance of existing structures and devices. To achieve this objective, accurate knowledge of the rollup mechanics that consider the effects of embedded heterogeneous units is necessary. In the present study, a comprehensive analytical model has been proposed to evaluate the dependence of self-rolling behaviors of strained membranes on the distribution and proportion of heterogeneous elements embedded, validated at macro and microscopic scales through molecular dynamic (MD) simulations and finite element (FE) simulations. In addition, our theoretical predictions combined with FE simulations could be used for precisely generating helical shapes with desired rolling curvatures, chirality, and helix angles, in excellent agreement with experimental observations, showing great potential in designing complex 3D structures based on strained membranes through incorporations of heterogeneous elements.

7.2 Introduction

Recently, significant research efforts have been focused on exploring and developing self-rolling technique aiming to achieve programmable shape and functional transformations based on bi and multilayers of strained membranes made from different materials and decorated with functional groups, which enables diverse applications including electro-mechanical/microelectromechanical systems (NEMS/MEMS) [1-3], sensors [4, 5], micro-robotics [6, 7], drug delivery [8, 9], and optoelectronics [10-12] as well as artificial biomimetic structures [13-20]. Particularly, more recent interest has been extended and triggered to incorporate new structural or compositional units into membranes and construct delicate inhomogeneous structures with tunable internal strain achieving a higher level of manipulation over the rollup geometry with enhanced performance [21-25]. For instance, alternative strips with varied chemical compositions have been utilized to construct the responsive material with local modulation of internal stress that enables controllable multiple 3D geometry transformations from selfbending to self-twisting [26]. Additionally, aligned carbon nanotubes (CNTs) in multiple directions have been reported to be embedded into the composite polymer membrane actuators to realize the accurate control of the self-rolling process and thus endow extra merits into the actuator, including high tensile strength along the fiber direction and electric conductivity [27]. It is noteworthy that one of the most significant feature of inhomogeneous strained structures is the directional self-rolling behaviors originating from anisotropic driving forces [28], offering flexible ways for constructing 3D architectures from predefined flat geometries. Recently, several theoretical models have been proposed to address the underlying mechanics of such morphologies transformation from 2D to 3D employed the continuum elasticity theory and differential geometry [28-30]. However, it remains quite challenging to analytically evaluate the anisotropic rolling curvatures of strained bilayer membranes with structural and

compositional inhomogeneity, and experimental measurements are still needed to obtain those values for the precise design of various 3D morphologies [28].

In the present study, we formulated a comprehensive analytical model to predict the self-rolling curvature of strained bilayer membranes with heterogeneous elements embedded, in which the contributions of both distribution and proportion of heterogeneous elements to the rollup curvature are accounted for. Our model predictions show the directional distribution of heterogeneous elements with different elastic modulus could induce anisotropic internal mismatch strain resulting in the anisotropic rollup behaviors, validated at macro and microscopic scales through molecular dynamic (MD) and FE simulations. In addition, as a demonstration of practical application for the incorporation of inhomogeneous structures, the FE simulations have been performed for generating helical shapes with desirable rolling curvatures and helix angles. The results are well agreed with experimental observations, showing great potential in designing complex 3D structures based on strained membranes through incorporation of heterogeneous elements.

7.3 Methodology

7.3.1 Analytical formulation

Motivated by the theory of laminated composites, our model considers a rectangular strained bilayer membranes of dimension L in length (x_1 direction) and W in width (x_2 direction), comprising an mixed active layer with aligned heterogeneous elements (top layer) and a passive matrix layer (bottom layer) bonded together as illustrated in Fig. 7.1. For sake of simplicity and without loss the generality, we only consider the aligned homogenous elements and assume the red and yellow parts in the Fig. 7.1 are the same kind of material, while blue

parts are introduced heterogeneous elements. The lattice constants of matrix and heterogeneous materials are expressed by a_s and a_f , respectively.



Figure 7.1. Schematic illustration of rectangular strained bilayer membranes with aligned heterogeneous elements colored in the blue.

The heterogeneous material, mixed layer and matrix are respectively of elastic constants $(C_{11f}, C_{12f}, C_{44f})$, $(C_{11f1}, C_{12f1}, C_{44f1})$, $(C_{11s}, C_{12s}, C_{44s})$, and thicknesses (h_f, h_{f1}, h_s) , where both materials are assumed to be cubic symmetrical. Here subscripts s, f and f_i denote the matrix, heterogeneous and mixed materials, respectively, and we continue to use this denotation in the following part. Then, the elastic modulus of mixed layer could be expressed using a weighted mean as

$$C_{11f1} = F \cdot C_{11f} + C_{11s} \cdot (1 - F) \tag{7.1}$$

$$C_{12f1} = F \cdot C_{12f} + C_{12s} \cdot (1 - F) \tag{7.2}$$

$$C_{44f1} = F \cdot C_{44f} + C_{44s} \cdot (1 - F) \tag{7.3}$$

Where F is the volume fraction of heterogeneous elements. The variation of internal strain induced by aligned heterogeneous elements is systematically examined using continuum elastic theory, and the deformation process of whole membranes is schematically illustrated in the Fig. 7.2. At the beginning (Fig. 7.2 (a)), the matrix and heterogeneous material are in a free state, and then they are bonded to each other after the heterogeneous elements are stretched to match the matrix (Fig. 7.2 (a)). Finally, they would contract and reach the equilibrium state. Here it is assumed that the matrix and heterogeneous elements must contract the same amount and obtain even outer surface in the equilibrium state, as depicted in Fig. 7.2 (c).



Figure 7.2. Exaggerated schematic pictures for the deformation process induced by integrating aligned heterogeneous elements (blue) to matrix material (yellow). (a) The matrix and heterogeneous material are in a free state. (b) The heterogeneous material is stretched and bonded with the matrix. (c) Two materials contract the same amount and obtain even outer surface in the equilibrium state.

Afterwards, the in plane anisotropic mismatch strains ($\varepsilon_{\parallel}, \varepsilon_{\perp}$), parallel and perpendicular to the direction of aligned heterogeneous elements are determined as following, based on the lattice constants of matrix and heterogeneous material.

$$\varepsilon_{\parallel} = \frac{c_{11s}F(c_{11f}^2 - c_{12f}^2)(a_s - a_f)}{a_f(c_{11f}^2 c_{11s}F - (c_{11s}^2 - c_{11s}^2)(F - 1)c_{11f} - c_{12f}^2 c_{11s}F)}$$
(7.4)

$$\varepsilon_{\perp} = \frac{a_s}{a_s + (a_s - a_f)F - M + N} - 1 \tag{7.5}$$

Coefficients M and N are given by

$$\boldsymbol{M} = \frac{c_{12f}a_f(c_{11s}^2 - c_{12s}^2)(a_s - a_f)(1 - F)F}{c_{11s}a_s(c_{11f}^2 - c_{12f}^2)F - c_{11f}(c_{11s}^2 - c_{12s}^2)(F - 1)a_f}$$
(7.6)

$$\boldsymbol{N} = \frac{C_{12s}a_s \left(C_{11f}^2 - C_{12f}^2\right) \left(a_s - a_f\right) (1 - F)F}{C_{11s}a_s \left(C_{11f}^2 - C_{12f}^2\right) F - C_{11f} \left(C_{11s}^2 - C_{12s}^2\right) (F - 1)a_f}$$
(7.7)

The relations of strain tensor $\varepsilon_{\alpha\beta}$ and displacement fields $u(u_1, u_2, u_3)$ of deformed membranes are evaluated in a given coordinate (x_1, x_2, x_3) based on the Von-Karman nonlinear theory describing the large transverse deflections of thin shell, as shown below,

$$\varepsilon_{\alpha\beta} = \frac{1}{2} \left(\frac{\partial u_{\alpha}}{\partial x_{\beta}} + \frac{\partial u_{\beta}}{\partial x_{\alpha}} + \frac{\partial u_{3}}{\partial x_{\beta}} \cdot \frac{\partial u_{3}}{\partial x_{\alpha}} \right) - x_{3} \frac{\partial^{2} u_{3}}{\partial x_{\alpha} \partial x_{\beta}}$$
(7.8)

where Greek subscripts α and β can have values 1 or 2. To use Rayleigh–Ritz method, the outof-plane displacement is assumed as

$$u_1(x_1, x_2) = \sum_{i=0}^3 \sum_{j=0}^3 A_{ij} x_1^i x_2^j , \qquad (7.9)$$

$$u_2(x_1, x_2) = \sum_{i=0}^3 \sum_{j=0}^3 B_{ij} x_1^i x_2^j, \qquad (7.10)$$

$$u_3 = \frac{1}{2}(ax_1^2 + bx_2^2), \qquad (7.11)$$

Where $(A_{ij}, B_{ij}, a, and b)$ are to-be-determined coefficients minimizing the potential energy U. Here the three-order polynomial approximation of in-plane displacement filed was proved to be sufficient to provide an accurate estimation for the self-rolling behaviors of strained bilayer membranes with inhomogeneous elements. And the three curvature fields of deformed membranes could be measured as following using transverse displacement

$$\kappa_{11} = \frac{\partial^2 u_3}{\partial x_1^2} \text{ and } \kappa_{22} = \frac{\partial^2 u_3}{\partial x_2^2}$$
(7.12)

Generally, the total strain energy U of the membranes follows by integrating the strain energy over the total volume as

$$U = \iiint_V \left(\frac{1}{2}C_{ijkl}^S \varepsilon_{kl}^S \varepsilon_{kl}^S + \frac{1}{2}C_{ijkl}^{f1} \varepsilon_{kl}^{f1} \varepsilon_{kl}^{f1}\right) dV$$
(7.13)

Where, C_{ijkl}^{S} and C_{ijkl}^{f1} are the fourth order elasticity tensors for matrix and mixed materials, separately. Subsequently the equilibrium configurations could be obtained via minimization of the total potential energy expression based on the trial in-plane displacement functions and transverse displacement shape functions. It is worth noted that in this quasi-static process, multi-stable equilibrium states have been detected using numerical methods, including the situations with two unequal final curvatures and with one principle curvature. We found the closed analytical solutions are only available for the case of one principle curvature, and fortunately, those configurations are exactly what those experiments [26, 27] have been observed.

$$\frac{1}{k_{11}} = \begin{bmatrix} \frac{(CF_1^3 - CF_1CF_2^2)h_f^5 + 5h_s(CF_1^2CS_1 - \frac{2}{5}CF_1CF_2CS_2 - \frac{3}{5}CF_2^2CS_1)h_f^4 + 6h_s^2(CF_1^2CS_1 + (\frac{2}{3}CS_1^2 - \frac{2}{3}CS_2^2)CF_1 - CF_2^2CS_1)h_f^3}{6h_s(h_s + h_f)h_f((CF_1^2 - CF_2^2)(CS_1\varepsilon_{\parallel} + CS_2\varepsilon_{\perp})h_f + h_s(CS_1^2 - CS_2^2)(CF_1\varepsilon_{\parallel} + CF_2\varepsilon_{\perp}))} + \frac{4h_s^3(CF_1^2CS_1 + (\frac{3}{2}CS_1^2 - \frac{3}{2}CS_2^2)CF_1 - CF_2^2CS_1)h_f^2 + 5h_s^4((CS_1^2 - \frac{3}{5}CS_2^2)CF_1 - \frac{2}{5}CF_2CS_1CS_2)h_f + (CS_1^3 - CS_1CS_2^2)h_s^5}{6h_s(h_s + h_f)h_f((CF_1^2 - CF_2^2)(CS_1\varepsilon_{\parallel} + CS_2\varepsilon_{\perp})h_f + h_s(CS_1^2 - CS_2^2)(CF_1\varepsilon_{\parallel} + CF_2\varepsilon_{\perp}))} \end{bmatrix}$$

$$\frac{1}{k_{22}} = \begin{bmatrix} \frac{(CF_1^3 - CF_1CF_2^2)h_f^5 + 5h_s(CF_1^2CS_1 - \frac{2}{5}CF_1CF_2CS_2 - \frac{3}{5}CF_2^2CS_1)h_f^4 + 6h_s^2(CF_1^2CS_1 + (\frac{2}{3}CS_1^2 - \frac{2}{3}CS_2^2)CF_1 - CF_2^2CS_1)h_f^3}{6h_s(h_s + h_f)h_f((CF_1^2 - CF_2^2)(CS_1\varepsilon_{\perp} + CS_2\varepsilon_{\parallel})h_f + h_s(CS_1^2 - CS_2^2)(CF_1\varepsilon_{\perp} + CF_2\varepsilon_{\parallel}))} + \frac{4h_s^3(CF_1^2CS_1 + (\frac{3}{2}CS_1^2 - \frac{2}{3}CS_2^2)CF_1 - CF_2^2CS_1)h_f^3}{6h_s(h_s + h_f)h_f((CF_1^2 - CF_2^2)(CS_1\varepsilon_{\perp} + CS_2\varepsilon_{\parallel})h_f + h_s(CS_1^2 - CS_2^2)(CF_1\varepsilon_{\perp} + CF_2\varepsilon_{\parallel}))} + \frac{4h_s^3(CF_1^2CS_1 + (\frac{3}{2}CS_1^2 - \frac{2}{3}CS_2^2)CF_1 - CF_2^2CS_1)h_f^3}{6h_s(h_s + h_f)h_f((CF_1^2 - CF_2^2)(CS_1\varepsilon_{\perp} + CS_2\varepsilon_{\parallel})h_f + h_s(CS_1^2 - CS_2^2)(CF_1\varepsilon_{\perp} + CF_2\varepsilon_{\parallel}))} \end{bmatrix}$$

$$(7.15)$$

Where
$$CF_1 = \frac{C_{11f_1}^2 - C_{12f_1} \cdot C_{12f_1}}{C_{11f_1}}$$
, $CF_2 = \frac{C_{12f_1} \cdot (C_{11f_1} - C_{12f_1})}{C_{11f_1}}$, $CS_1 = \frac{C_{12s}^2 - C_{12s}^2}{C_{11s}}$ and $CS_2 = \frac{C_{12s} \cdot (C_{11s} - C_{12s})}{C_{11s}}$.

7.3.2 Molecular dynamics simulations

As a generic example, the Cd-Te-S Stillinger-Weber potential[31] implemented using the LAMMPS package [32] was chosen to model the effects of inhomogeneity on self-rolling behaviors of CdSTe/CdTe systems. The interatomic potential, accurately predicts the lattice parameter, elastic constants and a wide variety of defect properties for bulk and multilayered thin crystals of II-VI semiconductor compounds, and has previously applied into the study of core/shell structured quantum dots [33], misfit dislocations[34], and heterogeneous film growth[35] for the Cd–Te–Se Ternary System. It is worth noting that the continuum model is applicable to any lattice-mismatched system, thus no particular material is required when comparing the MD and analytical results. Our calculations indicate that lattice constants of bulk crystals of CdS and CdTe are 5.835 Å and 6.478Å at 0 K, respectively, and the corresponding elastic constants are further evaluated using finite deformations methods, as shown in the Table 1, well agreed with previous results [31, 34].

Material	C ₁₁ (<i>GPa</i>)	C ₁₂ (<i>GPa</i>)	C44 (GPa)
CdTe	108	37.5	33
CdS	63.4	32	23

Table 7.1. The values of elastic constants used in the MD simulations

7.3.3 Numerical simulations

To avoid the time-consuming computation for MD simulations in large scale, finite element (FE) simulations were used for investigating the inhomogeneity induced competitive rollup behaviours and resulting helical geometries, implemented by the ABAQUS explicit/dynamic package [36]. The layered structures with predefined inhomogeneous strips were built in the module of composite layups, and 4-node doubly curved thin/thick shell elements with reduced integration (S4R) were used to discretize the shell geometry. The initial lattice mismatch strain was realized through setting different thermal expansion coefficients for each layer with controllable temperature variation. ABAQUS explicit/dynamic package was employed to examine the self-rolling behaviors of membranes with heterogeneous elements embedded.

7.4 Results and discussions

The predicted rolling curvatures of strained bilayer membranes with different distributions and volume fraction of heterogeneous elements determined by our analytical model are compared with results obtained by the MD simulations. Before considering the anisotropic roll-up behavior, we first simulated two isotropic self-rolling situations:1) no heterogeneous elements are introduced. 2) the heterogeneous elements are uniformly distributed in the top active layer. In these two cases, the rollup curvature radii along x_1 and x_2 direction would be equal $(\frac{1}{k_{11}} = \frac{1}{k_{22}})$ due to $\varepsilon_{\parallel} = \varepsilon_{\perp}$. The simulated geometry of the CdTe_xS₁x/CdTe system is shown in Fig. 7. 3 (a), where x_1 (perpendicular to the plane of paper), x_2 , and x_3 are aligned with [100], [010], and [001] respectively, and the periodic boundary condition is applied to x_1 or x_2 direction. The supercell is of dimensions 27.5 nm \times 109.7 nm along x_1 or x_2 direction, respectively, while for the x_3 direction, the thickness of CdTe matrix is fixed to be 3.24 nm, while that of active layer of CdTe_xS_{1-x} ranges from 4.54 to 9.7 nm. At the initial stage, the lattice of CdTe_xS_{1-x} will be stretched to match that of the matrix CdTe, as the driving force for self-rolling. After the relaxation, the rolling diameter is measured at the mid-plane of $CdTe_xS_{1-x}/CdTe$ system, through fitting and averaging the trajectories of high-energy atoms on the bottom and top surfaces.



Figure 7.3. The simulated geometry of the $CdTe_xS_{1-x}/CdTe$ system where Cd, Te and S atoms are colored red, blue and light yellow respectively. (b) The MD simulated rolling diameter as a function of the thickness for the self-rolling of CdS/CdTe and CdTe_0.5S_0.5/CdTe systems.

The MD simulated evolution of rollup diameter as a function of the thickness of active layer is presented in Fig. 7.3 (b), in comparison with the analytical predictions obtained from Eq (7.13-14). In the figure, x = 0 denotes the system of CdS/CdTe, while x = 0.5, as a generic example of uniformly distributed heterogeneous elements, represents the system of CdTe0.5S0.5/CdTe. It is indicated that the MD simulated values are in good agreement with data obtained from the analytical models. Additionally, it is also observed in the MD simulations that the anisotropic rollup behavior would not be triggered when the value of x is changed. The further validations are conducted by designing the alternative distributed strip structures for CdS/CdTe on the top layer, and a representative model with volume fraction F=0.6 is depicted in the Fig. 7.4.



Figure 7.4. The representative self-rolling models with alternative distributed strip structures of $CdTe_{0.4}S_{0.6}/CdTe$ (a) and (b) indicate the longitudinal and lateral alternative distributed strip structures, respectively, while (c) and (d) represent the corresponding equilibrium geometries.

The obtained results are illustrated in the Fig. 7.5, where the thickness of CdTe_xS1x/CdTe in Figs 7.5 (a) and (b), are set to be (3.24/3.24) nm and (3.24/4.54) nm, respectively, while, the dimensions along x_1 or x_2 direction remain the same as mentioned above. It reveals the effects of increased thickness and the varied volume fraction of active material CdS, and a good agreement has been achieved when *F* ranges from 0.3 to 0.9. We see that the predicted values deviate the simulated results when *F* is less than 0.3. It could be mainly attributed to the inaccurate anisotropic mismatch strain estimated previously, because our calculations are based on the assumption that when the matrix and heterogeneous elements are bonded together, they would contract the same amount and obtain even outer surface in the equilibrium state. However, it is no longer the real case, and an curved outer surface is expected in the equilibrium state, as shown in Fig. 7.6, when the volume fraction of active material is reduced. Our results reveal that the rolling diameter along longitudinal direction is always smaller than that of lateral direction, indicating that the longitudinal rolling has a lager curvature and would be the preferred direction, and this preference is strongly dependent on their respective elastic constants and volume fraction. It could be effectively predicted through our model and used to precise creating helical tubes.



Figure 7.5. The predicted and MD simulated rolling diameter as a function of the proportion (*F*) of active material CdS. In (a) and (b), the thickness of $CdTe_xS_{1-x}/CdTe$ are set to be (3.24/3.24) nm and (3.24/4.54) nm, respectively.



Figure 7.6. An curved outer surface would appear when the volume fraction of active material is reduced.

The further simulations for generating helical shapes are performed using FEM implemented by commercial software ABAQUS, considering that it is quite challenging and

time-consuming for MD simulations. The detailed rolling process for helical configurations are displayed in the Fig. 7.7, and the size on the rolling plane was reset to be a parallelogram with dimensions of 300 nm \times 3000 nm. The heterogeneous elements are initial embedded along the short edge of the parallelogram. The preferred rolling directions are perpendicular and parallel to the aligned direction of heterogeneous elements in Figs 7.7 (b) and (c), respectively. The elastic constants and thickness of CdTe_{0.5}S_{0.5}/CdTe used in Fig. 7.7 (b) are same as those used in Fig. 7.5 (a), but in Fig. 7.7 (c), the elastic constants of matrix and heterogeneous elements are exchanged to demonstrate the dependence of preference of rolling direction on their relative elastic constants.



Figure 7.7. FE simulated helical rolling processes of bilayer of $CdTe_{0.5}S_{0.5}/CdTe$ with alternative distributed strip structures. The initial geometry is shown in (a), where $CdTe_{0.5}S_{0.5}$ and CdTe are colored in red and blue, separately. The preferred rolling directions are perpendicular and parallel to the short edge of the parallelogram in (b) and (c), depending on the elastic constants of different strips, respectively.

7.5 Conclusion

In summary, in the present work, a theoretical model has been established to perform detailed investigations of the effects of inhomogeneity on the self-rolling curvature of strained bilayer membranes with verifications of MD simulations. The analytical predictions show good agreement with the MD simulated rollup diameter with both proportion of aligned heterogeneous elements and varied thickness of membranes considered. In addition, it has been revealed that the directional distribution of heterogeneous elements with different elastic modulus could induce anisotropic internal mismatch strain resulting in the anisotropic rollup curvatures. This implies one rolling direction would be always energetical preferred, and this preference strongly depends on the respective elastic constants of matrix and heterogeneous elements. The preferred rolling curvature and direction could be effectively predicted through our model and used to precise creating helical tubes using FEM simulation at macro scales. Further, our FEM simulations show the rolling process for generating helical configurations based on the analytical model, in excellent agreement with experimental observations, showing great potential in accurately designing complex 3D structures employing strained membranes through incorporations of heterogeneous elements.

7.6 Acknowledgement

We greatly thank the financial support from McGill Engineering Doctoral Award, China Scholarship Council, National Sciences and Engineering Research Council (NSERC) Discovery grant (grant # RGPIN-2017-05187), and NSERC Strategic grant (grant # STPGP 494012-16). We also acknowledge Supercomputer Consortium Laval UQAM McGill and Eastern Quebec for providing computing power.

7.7 Reference

[1] D.a. Grimm, Rolled-up nanomembranes as compact 3D architectures for field effect transistors and fluidic sensing applications, Nano Lett, 13 (2013) 213-218.

[2] O.G. Schmidt, C. Deneke, S. Kiravittaya, R. Songmuang, H. Heidemeyer, Y. Nakamura, R. Zapf-Gottwick, C. Muller, N.Y. Jin-Phillipp, Self-assembled nanoholes, lateral quantum-dot molecules, and rolled-up nanotubes, Ieee J Sel Top Quant, 8 (2002) 1025-1034.

[3] D.J. Bell, L. Dong, B.J. Nelson, M. Golling, Fabrication and Characterization of Three-Dimensional InGaAs GaAs Nanosprings, Nano, (2006).

[4] E.J. Smith, S. Schulze, S. Kiravittaya, Y. Mei, S. Sanchez, O.G. Schmidt, Lab-in-a-tube: Detection of individual mouse cells for analysis in flexible split-wall microtube resonator sensors, Nano Lett, 11 (2011) 4037-4042.

[5] I. Mnch, D. Makarov, R. Koseva, L. Baraban, D. Karnaushenko, C. Kaiser, K.F. Arndt, O.G. Schmidt, Rolled-up magnetic sensor: Nanomembrane architecture for in-flow detection of magnetic objects, ACS Nano, 5 (2011) 7436-7442.

[6] J.J. Abbott, K.E. Peyer, L.X.X. Dong, B.J. Nelson, How Should Microrobots Swim?, Springer Trac Adv Ro, 66 (2010) 157-167.

[7] J.J. Abbott, K.E. Peyer, M.C. Lagomarsino, L. Zhang, L.X. Dong, I.K. Kaliakatsos, B.J. Nelson, How Should Microrobots Swim?, Int J Robot Res, 28 (2009) 1434-1447.

[8] R. Arayanarakool, A.K. Meyer, L. Helbig, S. Sanchez, O.G. Schmidt, Tailoring threedimensional architectures by rolled-up nanotechnology for mimicking microvasculatures, Lab Chip, 15 (2015) 2981-2989.

[9] R. Fernandes, D.H. Gracias, Self-folding polymeric containers for encapsulation and delivery of drugs, Adv Drug Deliver Rev, 64 (2012) 1579-1589.

[10] S.M. Harazim, V.A.B. Quinones, S. Kiravittaya, S. Sanchez, O.G. Schmidt, Lab-in-a-tube: on-chip integration of glass optofluidic ring resonators for label-free sensing applications, Lab Chip, 12 (2012) 2649-2655.

[11] a. Bernardi, S. Kiravittaya, a. Rastelli, R. Songmuang, D.J. Thurmer, M. Benyoucef, O.G. Schmidt, On-chip Si/SiOx microtube refractometer, Appl Phys Lett, 93 (2008).

[12] S.M. Harazim, W. Xi, C.K. Schmidt, S. Sanchez, O.G. Schmidt, Fabrication and applications of large arrays of multifunctional rolled-up SiO/SiO₂ microtubes, Journal of Materials Chemistry, 22 (2012) 2878.

[13] K.U. Jeong, J.H. Jang, D.Y. Kim, C. Nah, J.H. Lee, M.H. Lee, H.J. Sun, C.L. Wang, S.Z.D. Cheng, E.L. Thomas, Three-dimensional actuators transformed from the programmed twodimensional structures via bending, twisting and folding mechanisms, Journal of Materials Chemistry, 21 (2011) 6824-6830.

[14] L. Ionov, Biomimetic Hydrogel-Based Actuating Systems, Advanced Functional Materials, 23 (2013) 4555-4570.

[15] L. Persano, A. Camposeo, D. Pisignano, Integrated bottom-up and top-down soft lithographies and microfabrication approaches to multifunctional polymers, J Mater Chem C, 1 (2013) 7663-7680.

[16] R. Kempaiah, Z.H. Nie, From nature to synthetic systems: shape transformation in soft materials, J Mater Chem B, 2 (2014) 2357-2368.

[17] M. Podgorski, D.P. Nair, S. Chatani, G. Berg, C.N. Bowman, Programmable Mechanically Assisted Geometric Deformations of Glassy Two-Stage Reactive Polymeric Materials, Acs Appl Mater Inter, 6 (2014) 6111-6119.

[18] A.R. Studart, Biologically Inspired Dynamic Material Systems, Angew Chem Int Edit, 54 (2015) 3400-3416.

[19] F.M. Wisser, B. Schumm, G. Mondin, J. Grothe, S. Kaskel, Precursor strategies for metallic nano- and micropatterns using soft lithography, J Mater Chem C, 3 (2015) 2717-2731.

[20] A.S. Gladman, E.A. Matsumoto, R.G. Nuzzo, L. Mahadevan, J.A. Lewis, Biomimetic 4D printing, Nat Mater, 15 (2016) 413-418.

[21] Q. Ge, H.J. Qi, M.L. Dunn, Active materials by four-dimension printing, Appl Phys Lett, 103 (2013).

[22] S. Janbaz, R. Hedayati, A.A. Zadpoor, Programming the shape-shifting of flat soft matter: from self-rolling/self-twisting materials to self-folding origami, Materials Horizons, 3 (2016) 536-547.

[23] Y.Q. Mao, Z. Ding, C. Yuan, S.G. Ai, M. Isakov, J.T. Wu, T.J. Wang, M.L. Dunn, H.J. Qi, 3D Printed Reversible Shape Changing Components with Stimuli Responsive Materials, Sci Rep-Uk, 6 (2016).

[24] J. Zhou, S.S. Sheiko, Reversible shape-shifting in polymeric materials, J Polym Sci Pol Phys, 54 (2016) 1365-1380.

[25] O. Kuksenok, A.C. Balazs, Stimuli-responsive behavior of composites integrating thermoresponsive gels with photo-responsive fibers, Materials Horizons, 3 (2016) 53-62.

[26] Z.L. Wu, M. Moshe, J. Greener, H. Therien-Aubin, Z.H. Nie, E. Sharon, E. Kumacheva, Three-dimensional shape transformations of hydrogel sheets induced by small-scale modulation of internal stresses, Nat Commun, 4 (2013).

[27] H. Lin, J. Gong, M. Eder, R. Schuetz, H. Peng, J.W.C. Dunlop, J. Yuan, Programmable Actuation of Porous Poly(Ionic Liquid) Membranes by Aligned Carbon Nanotubes, Advanced Materials Interfaces, (2016) 1600768.

[28] X.J. Yu, L.N. Zhang, N. Hu, H. Grover, S.C. Huang, D. Wang, Z. Chen, Shape formation of helical ribbons induced by material anisotropy, Appl Phys Lett, 110 (2017).

[29] Q. Guo, A.K. Mehta, M.A. Grover, W. Chen, D.G. Lynn, Z. Chen, Shape selection and multi-stability in helical ribbons, Appl Phys Lett, 104 (2014).

[30] Z. Chen, C. Majidi, D.J. Srolovitz, M. Haataja, Tunable helical ribbons, Appl Phys Lett, 98 (2011).

[31] X.W. Zhou, D.K. Ward, J.E. Martin, F.B. van Swol, J.L. Cruz-Campa, D. Zubia, Stillinger-Weber potential for the II-VI elements Zn-Cd-Hg-S-Se-Te, Phys Rev B, 88 (2013).

[32] S. Plimpton, Fast Parallel Algorithms for Short-Range Molecular-Dynamics, J Comput Phys, 117 (1995) 1-19.

[33] X.W. Zhou, M.E. Foster, F.B. van Swol, J.E. Martin, B.M. Wong, Analytical Bond-Order Potential for the Cd-Te-Se Ternary System, J Phys Chem C, 118 (2014) 20661-20679.

[34] X.W. Zhou, D.K. Ward, J.A. Zimmerman, J.L. Cruz-Campa, D. Zubia, J.E. Martin, F. van Swol, An atomistically validated continuum model for strain relaxation and misfit dislocation formation, J Mech Phys Solids, 91 (2016) 265-277.

[35] X.W. Zhou, J.J. Chavez, S. Almeida, D. Zubia, Understanding misfit strain releasing mechanisms via molecular dynamics simulations of CdTe growth on {112} zinc-blende CdS, J Appl Phys, 120 (2016).

[36] E.J. Barbero, Finite Element Analysis of Composite Materials using AbaqusTM, Taylor & Francis, 2013.

Chapter 8: Conclusions

8.1 Major conclusions and implications

This thesis provides a comprehensive body of work on the self-rolling behaviors of strain engineered nanomembranes accounting for several key aspects, i.e., lattice mismatch anisotropy, topological patterning, misfit dislocations, and compositional inhomogeneity. Predictive models have been developed to provide mechanistic guidance towards optimization and development of new microfabrication techniques of strain engineered nanomembranes with a higher level of manipulation over the size and geometry of the resultant roll-up structures. The major research findings of this thesis and their implication to practical applications are summarized below.

1. The effects of lattice mismatch strain anisotropy on multiple equilibrium states of the rolled-up nanomembrane have been investigated in detail.

Analytical expressions have been obtained for predicting the roll-up curvatures, validated through finite element (FE) simulations. The present study explicitly demonstrates the bidirectional nature of the self-rolling of nanomembranes, and clarifies the critical role of transverse strain in contributing to the discrepancy between previous model predictions and experimental measurements. Additionally, on base of our theoretical results, a couple of fabrication strategies, including third-layer deposition and corner geometry engineering, have been proposed to predictively manipulate the bidirectional rolling competition of strained nanomembranes, to achieve unidirectional roll-up. In particular, for the strategy of corner engineering, its practical applications have been verified through microfabrication experiments, demonstrating great effectiveness in improving the roll-up yield of microtubes.

Implication: This study provides useful guidelines for design and prediction of novel 3D nanomembrane geometries and suggest significant potential for optimization of the nanomembrane roll-up yield using the newly proposed fabrication strategies, i.e., third-layer deposition and corner geometry engineering.

2. A comprehensive model has been formulated to investigate the effects of surface patterning on self-rolling behaviors of strain engineered nanomembranes using von Karman nonlinear theory and Ritz method.

Our model predictions reveal that the size and density of patterns have significant influence on the final rollup curvature, validated through finite element (FE) simulations for the heteroepitaxial systems of GaAs/In_{0.2}Ga_{0.8}As and GaN/AlN. Our results explain the potential anisotropic self-rolling behaviors and preferred rollup direction originating from the directional distribution of surface patterns. In addition, the model is shown to be capable of achieving controllable helical rolling of nanomembranes with desired helical angle and pitch period as well as diameter D through strip patterning, in excellent agreement with experimental observations.

Implication: The findings can directly translate into valuable design insights for more complex roll-up geometries, such as helical structures with desired geometric metrics (i.e., helical angle, pitch period and diameter).

3. A combined framework that connects the atomic length scale variations of dislocations to continuum elasticity, has been presented to quantitatively evaluate the effects of misfit dislocations on the self-rolling of strain-engineered nanomembranes.

The typical misfit dislocations of (GaN/In_{0.5}Ga_{0.5}N) with wurtzite lattice, and (CdTe/CdTe_{0.5}S_{0.5}) with zinc-blend lattice have been created in MD simulations and their generalized stacking fault energy curves are calculated to be an input of Peierls-Nabarro model to obtain the disregistry functions of dislocations that accurately determine the strain relaxation effects induced by

147

lattice elastic distortion of dislocations. The potential influences of misfit dislocations on the self-rolling behaviors of strain-engineered nanomembranes are further investigated employing von Karman shell theory with the disregistry functions incorporated. It is found that, besides the influences of elastic anisotropy of materials, the formation of misfit dislocations could be another potential contributing factor causing the anisotropic rolling of strain-engineered nanomembranes. Additionally, we can see that independent of the thickness, the increased dislocation density would enhance the rollup diameters of released nanomembranes. The theoretical predictions are confirmed by the MD simulations of rolling up of nanomembranes with the existence of misfit dislocations, and the analytical method is shown to be reliable when applicable into different crystal geometries and dislocation configurations.

Implication: The results provide effective ways to predict and design the rollup configurations of strain-engineered semiconductor nanomembrane containing misfit dislocations with a relatively large mismatch strain and thickness.

4. A theoretical model has been established to perform detailed investigations of effects of inhomogeneity on the self-rolling curvature of strained bilayer membranes with verifications of MD simulations.

The analytical predictions show well agreement with the MD simulated rollup diameter with varied thickness and heterogeneous proportion of membranes. Meanwhile, it has been revealed that the directional distribution of heterogeneous elements with different elastic modulus could induce anisotropic internal mismatch strain resulting in the anisotropic rollup curvatures. This implies one rolling direction would be always energetical preferred, and this preference strongly depends on the respective elastic constants of matrix and heterogeneous elements. The preferred rolling curvature and direction could be effectively predicted through our model and used to precise creating helical configuration using FE simulations.

Implication: the present study suggests great potential in accurately designing complex 3D

structures employing strained membranes through incorporations of heterogeneous elements. More application potentials related to the incorporation of heterogeneous elements are expected to be explored in the future.

8.2 Contribution to the original knowledge

The contributions to the original knowledge are enlisted as follows:

a) The effects of lattice mismatch strain anisotropy on the self-rolling of nanomembranes have been first clarified, and the critical role of transverse strain in contributing to the discrepancy between previous model predictions and experimental measurements has been quantified, for which no systematic study is reported in the literature,

b) The new microfabrication strategies including third-layer deposition and corner geometry engineering, have been proposed to predictively manipulate the bidirectional rolling competition of strained nanomembranes, suggest significant potential for optimization of the nanomembrane roll-up yield.

c) The dependence of rollup direction and curvature of rollup nanomembranes on the size and density of patterns has been systematically studied for the first time, providing critical insights for the design of complex 3D structures of self-assembly of nanomembranes through controllable surface patterning.

d) The strain relaxation effects induced by misfit dislocation on the self-rolling of strainengineered nanomembranes have been quantified for the first time, which helps precisely design the rollup structures of nanomembranes with the consideration of dislocations especially for the situations of large mismatched strain and thickness, and reduce the deviations of theoretical prediction for experimental observations.

e) A comprehensive theoretical framework has been proposed to investigate the effects of heterogeneous elements on the self-rolling behaviors, which is not yet available in the literature.

149

It shows great potential in designing geometrically complex 3D shell structures employing strained membranes through incorporation of heterogeneous elements;

8.3 Future work

Potential directions for future work beyond this study are summarized below:

1. Effects of initial curvatures on the self-rolling of nanomembranes

Previous studies of self-rollup behaviors of nanomembranes mainly focusses on the fabrications of initially flat nanomembranes on regular substrates, and only tubular architectures with single direction curvature are favorable. Both fabrication and investigation of three-dimensional architectures with adjustable bidirectional curvature remains a huge challenging. It is therefore important to identify the effects of initial Gaussian curvature of nanomembranes on the resulting self-rolling structures. The investigation is expected to contribute to the multiple equilibrium configurations with bidirectional curvature [1-3] and provide possibilities to reveal the curvature-induced surface effect at small dimensions [4-8].

2. Multiple external signals triggered programmable self-rolling/ self-twisting for various active materials.

This thesis mainly focuses on the mismatch strain triggered self-rolling process, but other external signals (like magnetic field, temperature, swelling and pH) are not involved [9]. It is expected that more active materials responsive to various external signal could be incorporated into strain engineered nanomembranes to produce bending/rolling, which could produce extra design flexibility for the rollup of multilayer structures and realize time dependent configurations (called as 4D structures) [10, 11].

3. Synergistic effects of constituent materials on the rolled up nanomembranes.

The concept of mechanical self-rolling of nanomembranes, initially developed for fabricating semiconductor, has been recently extended to the production of hybrid metal-semiconductor structures [12], metal nanomembranes [13], organic Nano shells [14] and metal-films formed

from strained layers deposited onto polymer films [5]. There has been a clear tendency using multi-materials system to fabricate the biomedical devices [15, 16], robotics [17] or traditional MEMS/NEMS applications but achieving higher performance [18]. Much future work is still required to explore the synergistic effects of constituent materials on the rolled up nanomembranes.

4. Quantitively analysis and predictive design of Kiri-origami microstructures based on the self-rolling technology.

The self-rolling technology has also been expanded to fabricate Kiri-origami microstructures, which has made possible the creation of an enormous amount of new designs, and enable multistability that allows reprogrammable reconfigurations [19-22]. Moreover, appropriate software package based on origami mathematics has been available and ready to solve the inverse origami problems and design significantly complex 3D origami geometry [23]. Despite the fundamental geometry and elastic theory of origami-based metamaterials have been addressed [22, 24], several issues need to be further discussed and improved with respect to the self-rolling behaviors of connected elements in those origami patterns, especially for the effects of geometrical and mechanical parameters including the thickness, elastic constants, temperature induced strain gradient, and the controlled actuation sequence [25].

5. Integration of optoelectronic and biomedical modules into rolled-up geometry.

The integration of diverse heterogeneous materials on the self-rollup geometry would allow optoelectronic and biomedical functionalities. In these systems, the curvature-dependent spatial strain distribution needs to be elaborately designed avoiding cracking or delamination after self rolling process [26]. Therefore, multiscale methods need to be further developed and applied to reveal the self rolling mechanism of these complex materials and to optimize functionalities and enhance the performance.

8.4 References

[1] E. Eckstein, A. Pirrera, P.M. Weaver, Multi-mode morphing using initially curved composite plates, Compos Struct, 109 (2014) 240-245.

[2] L. Giomi, L. Mahadevan, Multi-stability of free spontaneously curved anisotropic strips, P Roy Soc a-Math Phy, 468 (2012) 511-530.

[3] S. Vidoli, C. Maurini, Tristability of thin orthotropic shells with uniform initial curvature, P Roy Soc a-Math Phy, 464 (2008) 2949-2966.

[4] O.V. Pylypovskyi, V.P. Kravchuk, D.D. Sheka, D. Makarov, O.G. Schmidt, Y. Gaididei, Coupling of Chiralities in Spin and Physical Spaces: The Mobius Ring as a Case Study, Phys Rev Lett, 114 (2015).

[5] Y.F. Mei, G.S. Huang, A.A. Solovev, E.B. Urena, I. Moench, F. Ding, T. Reindl, R.K.Y. Fu, P.K. Chu, O.G. Schmidt, Versatile Approach for Integrative and Functionalized Tubes by Strain Engineering of Nanomembranes on Polymers, Adv Mater, 20 (2008) 4085-4090.

[6] T. Huang, Z.Q. Liu, G.S. Huang, R. Liu, Y.F. Mei, Grating-structured metallic microsprings, Nanoscale, 6 (2014) 9428-9435.

[7] R. Streubel, L.Y. Han, F. Kronast, A.A. Unal, O.G. Schmidt, D. Makarov, Imaging of Buried 3D Magnetic Rolled-up Nanonnembranes, Nano Lett, 14 (2014) 3981-3986.

[8] R. Streubel, D.J. Thurmer, D. Makarov, F. Kronast, T. Kosub, V. Kravchuk, D.D. Sheka, Y. Gaididei, R. Schafer, O.G. Schmidt, Magnetically Capped Rolled-up Nanomembranes, Nano Lett, 12 (2012) 3961-3966.

[9] W. Huang, X. Yu, P. Froeter, R.M. Xu, P. Ferreira, X.L. Li, On-Chip Inductors with Self-Rolled-Up SiNx Nanomembrane Tubes: A Novel Design Platform for Extreme Miniaturization, Nano Lett, 12 (2012) 6283-6288.

[10] T.G. Leong, C.L. Randall, B.R. Benson, N. Bassik, G.M. Stern, D.H. Gracias, Tetherless thermobiochemically actuated microgrippers, P Natl Acad Sci USA, 106 (2009) 703-708.

[11] A.S. Gladman, E.A. Matsumoto, R.G. Nuzzo, L. Mahadevan, J.A. Lewis, Biomimetic 4D printing, Nat Mater, 15 (2016) 413-418.

[12] V.Y. Prinz, S.V. Golod, Elastic Silicon-Film-Based Nanoshells: Formation, Properties, and Applications, J Appl Mech Tech, 47 (2006) 867-878.

[13] Y.V. Nastaushev, V.Y. Prinz, S.N. Svitasheva, A technique for fabricating Au/Ti microand nanotubes, Nanotechnology, 16 (2005) 908-912.

[14] V. Luchnikov, O. Sydorenko, M. Stamm, Self-rolled polymer and composite polymer/metal micro- and nanotubes with patterned inner walls, Adv Mater, 17 (2005) 1177-1182.

[15] M. Jamal, A.M. Zarafshar, D.H. Gracias, Differentially photo-crosslinked polymers enable self-assembling microfluidics, Nat Commun, 2 (2011).

[16] K.U. Jeong, J.H. Jang, D.Y. Kim, C. Nah, J.H. Lee, M.H. Lee, H.J. Sun, C.L. Wang, S.Z.D. Cheng, E.L. Thomas, Three-dimensional actuators transformed from the programmed twodimensional structures via bending, twisting and folding mechanisms, Journal of Materials Chemistry, 21 (2011) 6824-6830.

[17] H.W. Huang, M.S. Sakar, A.J. Petruska, S. Pane, B.J. Nelson, Soft micromachines with programmable motility and morphology, Nat Commun, 7 (2016).

[18] C. Liu, Recent developments in polymer MEMS, Adv Mater, 19 (2007) 3783-3790.

[19] J.L. Silverberg, A.A. Evans, L. McLeod, R.C. Hayward, T. Hull, C.D. Santangelo, I. Cohen, Using origami design principles to fold reprogrammable mechanical metamaterials, Science, 345 (2014) 647-650.

[20] F. Lechenault, M. Adda-Bedia, Generic Bistability in Creased Conical Surfaces, Phys Rev Lett, 115 (2015).

[21] J.L. Silverberg, J.H. Na, A.A. Evans, B. Liu, T.C. Hull, C.D. Santangelo, R.J. Lang, R.C. Hayward, I. Cohen, Origami structures with a critical transition to bistability arising from hidden degrees of freedom (vol 14, pg 389, 2015), Nat Mater, 14 (2015) 540-540.

[22] S. Waitukaitis, R. Menaut, B.G.G. Chen, M. van Hecke, Origami Multistability: From Single Vertices to Metasheets, Phys Rev Lett, 114 (2015).

[23] J. Rogers, Y.G. Huang, O.G. Schmidt, D.H. Gracias, Origami MEMS and NEMS, Mrs Bull, 41 (2016) 123-129.

[24] V. Brunck, F. Lechenault, A. Reid, M. Adda-Bedia, Elastic theory of origami-based metamaterials, Phys Rev E, 93 (2016).

[25] N. An, M.E. Li, J.X. Zhou, Predicting origami-inspired programmable self-folding of hydrogel trilayers, Smart Mater Struct, 25 (2016).

[26] V.B. Shenoy, D.H. Gracias, Self-folding thin-film materials: From nanopolyhedra to graphene origami, Mrs Bull, 37 (2012) 847-854.