STUDY OF DYNAMICS AT A SOFT INTERFACE: THE ROLE OF GEOMETRY, FRICTION, AND SURFACE ENERGY

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STUDY OF DYNAMICS AT A SOFT INTERFACE: THE ROLE OF GEOMETRY, FRICTION, AND SURFACE ENERGY

by

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Submitted

in fulfillment of requirements of degree of *Doctor of Philosophy*

to the



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To God, with gratitude for His blessings

&

To my parents, Asha Chawla and
Surinder Chawla

Certificate

This is to certify that the thesis titled **"Study of dynamics at a soft interface: the role of geometry, friction, and surface energy"** *being submitted by* **Ms. Aashna** *to the Indian Institute of Technology Delhi for the award of the degree of* **Doctor of Philosophy** *in Physics is a record of bonafide research work carried out by her under my supervision and guidance. She has fulfilled the requirements for the submission of the thesis, which to the best of my knowledge has reached the required standard.*

The material contained in this thesis has not been submitted in part or full to any other University or Institute for the award of any degree or diploma.



06-07-2026

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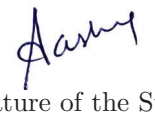
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Aashna

Abstract

Interfaces, formed when two or more phases come together, display interesting properties and are ubiquitous in nature. A unique property of interfaces is the existence of interfacial stresses. While interfacial stresses are usually negligible in rigid solids, they play a significant role in soft and biological systems. The relative importance of interfacial stresses and elasticity in soft solids depends on the length scale being probed. Many interesting previous works have studied the interplay between surface stresses and elasticity at a static soft interface and how it gives rise to a diverse range of patterns at the interface. In this thesis, we examine the nature of dynamics at a soft interface and the phenomena that emerge from the coupling of dynamics with elasticity and interfacial stresses. We design a model interface consisting of a highly bendable thin polymer sheet that is weakly adhered to a water-swollen hydrogel substrate to study the dynamics and discover some interesting new phenomenology.

An important feature of dynamics at an interface is the nature and role of friction. Conventional techniques for measuring friction typically involve point or planar contacts. However, real soft and biological systems come in a variety of shapes and geometries. We develop an experimental system that enables us to measure friction between two soft surfaces of different intrinsic shapes moving relative to each other- thereby functioning as a sensitive tribometer. We examine how geometry affects friction at this thin sheet-hydrogel interface. We find a strong dependence of friction on the relative geometry of the two surfaces: a flat sheet experiences significantly larger friction on a spherical substrate than on a flat or cylindrical substrate. We show that the stress developed in the sheet

due to its geometrically incompatible confinement is responsible for the enhanced friction. This mechanism also shows a size-dependent friction with a transition in the nature of friction as the sheet radius is increased beyond a critical value. Our finding reveals a hitherto unnoticed mechanism based on an interplay between geometry and elasticity that may influence friction significantly in soft, biological, and nanoscale systems. In particular, it provokes us to reexamine our understanding of phenomena such as the curvature dependence of the mobility of biological cells.

We use the compressive stress arising from the frictional interaction between the sheet and the hydrogel substrate in the above-described experimental system to slowly and controllably crumple extremely thin sheets and study their crumpling dynamics. Through these controlled crumpling experiments on thin sheets, we find a new class of elastocapillary phenomenon, which we refer to as the capillary crumpling transition. We find that as we begin to compress the sheet, initially it becomes increasingly difficult to compress the sheet further; however, after a well-defined critical compression, the sheet transitions into a self-crumpling state. The transition observed by us reminds one of the crumpling transition that has been theoretically predicted in thermalized sheets at temperatures above a critical threshold, but has never been convincingly observed experimentally. We show that the capillary crumpling transition we observed is driven by the sheet's surface energy. A comparison of the relative importance of surface and elastic energy in a typical crumpling process allows us to introduce a dimensionless parameter, the capillary crumpling number, that measures the sheet's tendency to undergo the capillary crumpling transition. We found that many-layered biological tissues and two-dimensional nanomaterials fall within the regime of a large capillary crumpling

number. The transition is characterized by the percolation of a fold network and a power-law increase in fold density, typical signatures of a critical phenomenon.

In our experiments, the crumpled state of the sheet displays a tunable order of folds, establishing the phenomenon's potential as a simple and scalable technique to do origami with extremely thin sheets. We can tune the symmetry of the fold pattern in the sheet using the substrate geometry. We also use a pattern of grooves engraved on a flat hydrogel substrate to imprint desired fold pattern in the sheet. The technique further allows us to mold a 2D flat sheet into a 3D shape. The 3D shape is maintained even after the sheet is released from the substrate. We also show the ability to uncrumple and retrieve the flat sheet by using the force of surface tension of water. The resulting flat sheet preserves the pattern of creases reminiscent of the fold patterns that were imprinted on it, which are expected to modulate the mechanical response of the sheet.

सार

जब दो या अधिक अवस्थाएँ एक साथ आती हैं, तो जो सीमाएँ (इंटरफेस) बनती हैं, वे रोचक गुण प्रदर्शित करती हैं और प्रकृति में सर्वत्र पाई जाती हैं। इंटरफेस का एक विशिष्ट गुण अंतरफलीय तनावों का अस्तित्व है। जहाँ कठोर ठोस पदार्थों में अंतरफलीय तनाव सामान्यतः नगण्य होते हैं, वहीं मुलायम और जैविक प्रणालियों में ये महत्वपूर्ण भूमिका निभाते हैं। मुलायम ठोसों में अंतरफलीय तनाव और प्रत्यास्थता (इलास्टिसिटी) का सापेक्ष महत्व उस लंबाई पैमाने पर निर्भर करता है जिसकी जाँच की जा रही हो। पूर्व के अनेक रोचक अध्ययनों में स्थिर मुलायम इंटरफेस पर सतही तनाव और प्रत्यास्थता के पारस्परिक प्रभाव तथा उससे उत्पन्न विभिन्न प्रकार के पैटर्न और घटनाओं का अध्ययन किया गया है। इस शोध–प्रबंध में हम एक मुलायम इंटरफेस की गतिशील प्रकृति और गतिशीलता, प्रत्यास्थता तथा अंतरफलीय तनावों के युग्मन से उत्पन्न होने वाली घटनाओं की जाँच करते हैं। इसके लिए हम एक मॉडल इंटरफेस तैयार करते हैं, जिसमें एक अत्यधिक लचीली पतली पॉलिमर शीट को पानी से फूले हुए हाइड्रोजेल सब्सट्रेट से हल्के रूप से चिपकाया जाता है, ताकि हम उसकी गतिशीलता का अध्ययन कर सकें और कुछ नई रोचक घटनाओं की खोज कर सकें।

इंटरफेस पर गतिशीलता का एक महत्वपूर्ण पहलू घर्षण की प्रकृति और भूमिका है। घर्षण मापने की पारंपरिक तकनीकें सामान्यतः बिंदु या समतलीय संपर्कों पर आधारित होती हैं। किंतु वास्तविक मुलायम और जैविक प्रणालियाँ विभिन्न आकृतियों और ज्यामितियों में पाई जाती हैं। हम एक ऐसी प्रायोगिक प्रणाली विकसित करते हैं जो हमें भिन्न आंतरिक आकृतियों वाली दो मुलायम सतहों के बीच, उनके आपसी सापेक्ष गति के दौरान, घर्षण को मापने में सक्षम बनाती है—इस प्रकार यह एक संवेदनशील ट्राइबोमीटर के रूप में कार्य करती है। हम इस पतली शीट–हाइड्रोजेल इंटरफेस पर ज्यामिति के घर्षण पर प्रभाव की जाँच करते हैं। हम पाते हैं कि घर्षण दो सतहों की सापेक्ष ज्यामिति पर दृढ़ता से निर्भर करता है: एक समतल शीट को गोलाकार सब्सट्रेट पर समतल या बेलनाकार सब्सट्रेट की तुलना में कहीं अधिक घर्षण का अनुभव होता है। हम दिखाते हैं कि शीट में उत्पन्न तनाव, जो उसकी ज्यामितीय रूप से असंगत बंदीकरण के कारण विकसित होता है, बढ़े हुए घर्षण के लिए जिम्मेदार है। यह तंत्र

आकार–निर्भर घर्षण भी दर्शाता है, जिसमें शीट की त्रिज्या एक निश्चित महत्वपूर्ण मान से अधिक होने पर घर्षण की प्रकृति में परिवर्तन होता है। हमारा निष्कर्ष ज्यामिति और प्रत्यास्थता के पारस्परिक प्रभाव पर आधारित एक अब तक अनदेखे तंत्र को उजागर करता है, जो मुलायम, जैविक और नैनोस्तरीय प्रणालियों में घर्षण को महत्वपूर्ण रूप से प्रभावित कर सकता है। विशेष रूप से, यह हमें जैविक कोशिकाओं की गतिशीलता की वक्रता–निर्भरता जैसे घटनाक्रमों की अपनी समझ पर पुनर्विचार करने के लिए प्रेरित करता है।

उपरोक्त वर्णित प्रायोगिक प्रणाली में शीट और हाइड्रोजेल सब्सट्रेट के बीच घर्षणीय अंतःक्रिया से उत्पन्न संपीडन तनाव का उपयोग हम अत्यंत पतली शीटों को धीरे और नियंत्रित रूप से मरोड़ने (क्रम्पल करने) के लिए करते हैं और उनकी क्रम्पलिंग गतिकी का अध्ययन करते हैं। इन नियंत्रित प्रयोगों के माध्यम से हम एक नई श्रेणी की इलास्टोकैपिलरी घटना पाते हैं, जिसे हम "कैपिलरी क्रम्पलिंग संक्रमण" कहते हैं। हम पाते हैं कि जब हम शीट को संपीडित करना प्रारंभ करते हैं, तो प्रारंभ में उसे और अधिक संपीडित करना क्रमशः कठिन होता जाता है; किंतु एक स्पष्ट रूप से परिभाषित महत्वपूर्ण संपीडन के बाद शीट स्वयं–क्रम्पल अवस्था में चली जाती है। हमारे द्वारा देखा गया यह संक्रमण उस क्रम्पलिंग संक्रमण की याद दिलाता है जिसकी सैद्धांतिक भविष्यवाणी तापीयीकृत शीटों में एक महत्वपूर्ण तापमान सीमा से ऊपर की गई थी, किंतु जिसे अब तक प्रयोगात्मक रूप से विश्वसनीय रूप से नहीं देखा गया था। हम दिखाते हैं कि हमारे द्वारा देखा गया कैपिलरी क्रम्पलिंग संक्रमण शीट की सतही ऊर्जा द्वारा प्रेरित होता है। एक सामान्य क्रम्पलिंग प्रक्रिया में सतही और प्रत्यास्थ ऊर्जा के सापेक्ष महत्व की तुलना हमें एक आयामहीन पैरामीटर प्रस्तुत करने की अनुमति देती है, जिसे हम "कैपिलरी क्रम्पलिंग संख्या" कहते हैं, जो शीट की इस संक्रमण से गुजरने की प्रवृत्ति को मापती है। हमने पाया कि बहु–परत जैविक ऊतक और द्वि–आयामी नैनो–सामग्री बड़ी कैपिलरी क्रम्पलिंग संख्या वाले क्षेत्र में आती हैं। यह संक्रमण मोड़ों के नेटवर्क के परकोलेशन और मोड़ घनत्व में घातीय नियम (पावर–लॉ) वृद्धि द्वारा चिह्नित होता है, जो किसी महत्वपूर्ण (क्रिटिकल) घटना के विशिष्ट संकेत हैं।

हमारे प्रयोगों में शीट की क्रम्पल अवस्था मोड़ों के क्रम को समायोज्य (ट्यून करने योग्य) रूप में प्रदर्शित करती है, जिससे यह घटना अत्यंत पतली शीटों के साथ ओरिगामी करने की एक सरल और मापनीय तकनीक के रूप में संभावनाएँ स्थापित करती है। हम सब्सट्रेट की ज्यामिति का उपयोग

करके शीट में मोड़ पैटर्न की सममिति को नियंत्रित कर सकते हैं। हम समतल हाइड्रोजेल सब्सट्रेट पर उकेरी गई खाँचों की एक रूपरेखा का उपयोग करके शीट में इच्छित मोड़ पैटर्न अंकित भी करते हैं। यह तकनीक एक द्वि–आयामी समतल शीट को त्रि–आयामी आकृति में ढालने की भी अनुमति देती है। शीट को सब्सट्रेट से मुक्त करने के बाद भी यह त्रि–आयामी आकृति बनी रहती है। हम यह भी दिखाते हैं कि पानी के पृष्ठ तनाव की शक्ति का उपयोग करके शीट को फिर से खोलकर समतल अवस्था में लौटाया जा सकता है। परिणामी समतल शीट पर उन मोड़ पैटर्नों की स्मृति स्वरूप सिलवटें बनी रहती हैं जो उसमें अंकित की गई थीं, और ये शीट की यांत्रिक प्रतिक्रिया को संशोधित करने की अपेक्षा रखती हैं।

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List of Symbols

γ	Surface energy
A	Area
Υ	Surface stress
p	pressure
κ	Curvature
ϵ	Strain
σ	Stress
E	Young's Modulus
F_{fr}	Friction force
μ	Coefficient of friction
N	Normal load
M	Mass of block
g	Acceleration due to gravity
ℓ_{ec}	Elastocapillary length
ℓ_{bc}	Bendocapillary length
λ	Wavelength of wrinkles
h	Thickness of polymer thin sheet

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Y	Stretching Modulus of thin sheet
B	Bending Modulus of thin sheet
ν	Poisson's ratio
$\mathcal{U}$	Energy density (per unit area)
U	Energy
R	Radius of hydrogel substrate
R_0	Initial radius of hydrogel substrate
w	Projected sheet radius
W	Radius of sheet
W_0	Initial radius of sheet
$\tilde{R}$	Normalized substrate radius
$\tilde{W}$	Normalized sheet's radius
f	Fold density
L	Length of largest fold
ℓ	Normalized length of largest fold
v	Relative velocity
Γ	Strength of frictional interaction between sheet and hydrogel substrate
m	Limiting value of slope of $\tilde{W}(\tilde{R})$ curve as $\tilde{R} \to 1$
χ	Slope of $\tilde{W}(\tilde{R})$ curve
$(\tilde{R}_c, \tilde{W}_c)$	Transition point
t	Time
t_w	Time after which the experiment is stopped
k_B	Boltzmann constant
T	Temperature

X	Ridge length
Cr	Capillary crumpling number