



# Spontaneous instability of soft thin films on curved substrates due to van der Waals interaction

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## ABSTRACT

The linear bifurcation theory is used to investigate the stability of soft thin films bonded to curved substrates. It is found that such a film can spontaneously lose its stability due to van der Waals or electrostatic interaction when its thickness reduces to the order of microns or nanometers. We first present the generic method for analyzing the surface stability of a thin film interacting with the substrate and then discuss several important geometric configurations with either a positive or negative mean curvature. The critical conditions for the onset of spontaneous instability in these representative examples are established analytically. Besides the surface energy and Poisson's ratio of the thin film, the curvature of the substrate is demonstrated to have a significant influence on the wrinkling behavior of the film. The results suggest that one may fabricate nanopatterns or enhance the surface stability of soft thin films on curved solid surfaces by modulating the mechanical properties of the films and/or such geometrical properties as film thickness and substrate curvature. This study can also help to understand various phenomena associated with surface instability.

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## 1. Introduction

Surface instability of thin solid films is a crucial issue for many technologically important applications. Experimental observations find that under certain conditions, irregular roughness or regular patterns may form on the surfaces of free or stressed thin films. On one hand, thin films are often required in many engineering fields to have a stable and smooth surface morphology and various techniques have been developed to prevent surface wrinkling or roughening of materials. On the other hand, surface microstructures consisting of regular patterns endow the materials with some unusual properties and novel applications. For example, introducing micro- or nanosized surface structures can make a solid material to have the superhydrophobic and self-cleaning property, and special optical features can be rendered by fabricating regular distributed quantum dots with a characteristic wavelength (Gao and Nix, 1999). Therefore, controlled surface patterning of thin films hold great promise for a wide range of industrial applications in, for instance, metallurgy, microelectronics, stretchable electronic interconnects and devices, diffraction gratings, lithography, and characterization of the mechanical properties of thin films. Considerable efforts have been directed towards elucidating the physical mechanisms and pattern characteristics of surface instability (Spencer et al., 1991; Gao, 1994; Kukta and Freund, 1997; Lu and Suo, 2001, 2002; Chen and Hutchinson, 2004; Harrison et al., 2004; Stafford et al., 2004; Huang et al., 2005; Huang, 2005; Khang et al., 2006; Audoly and Boudaoud, 2008; Jiang et al., 2008; Im and Huang, 2008; Li et al., 2010). In addition, surface wrinkling also plays a critical role in some

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biophysical processes and biological functions in animals and plants (Cerda and Mahadevan, 2003; Efimenko et al., 2005; Genzer and Groenewold, 2006; Feng et al., 2007; Li et al., 2011).

The issue of the surface wrinkling of soft polymer films (e.g., with shear modulus smaller than 10 MPa) is of special significance in the fields of lithography, organic electronics, and biochips (Chou et al., 1999; Huang et al., 2006, 2008) and also has invoked increasing interest. Owing to their sensitivity to external stimuli, soft thin films can be destabilized by such interactions as van der Waals force, electrostatic force, and capillary force (Shenoy and Sharma, 2001, 2002; Wang et al., 2005; Yoon et al., 2005; Huang et al., 2007). For example, when a rigid contactor or planar indenter approaches normally a soft elastic thin film lying on a substrate, the film surface can be destabilized by the van der Waals interaction between the contactor and the film. If the surface energy of the film is negligible, the induced pattern wavelength is proportional to the film thickness but independent of the nature of the interaction force. When the film thickness reduces to less than 1  $\mu\text{m}$ , the dependence relationship between the wavelength and the film thickness becomes nonlinear due to the enhanced contribution of surface energy. Similar phenomena have been reported for soft elastic films in an electrostatic field (Sarkar et al., 2008). This result for thin elastic films differs from that for viscous liquid films (Herminghaus, 1999; Schäffer et al., 2000), in which the wavelength and pattern characteristics depend strongly on the nature and magnitude of the external stimuli.

Most previous studies have been focused on the surface instability and pattern formation of originally planar films. Due to the effects of topological constraint and surface tension, however, the wrinkling of curved surfaces exhibits some features distinct from those of planar surfaces. Surface patterns on soft matter with curved geometry arise in a large number of physical and biological applications (Li et al., 2009; Ben Amar and Ciarletta, 2010). For instance, the surface morphology of colloidal particles can be utilized as a suitable intermediate to grow a metal, semiconductor, or quantum dot layer on a silica shell (Ye et al., 2008). During the possessing of polymer fibers, wrinkling surfaces are often observable (Wu et al., 2008; Wang et al., 2009). Surface buckling and the induced patterns also play significant roles in some biological functions and processes of tissues and organs both in animals and plants (Ben Amar and Goriely, 2005; Boudaoud, 2010). For example, a growing tumor may keep a spherical shape or bifurcate to a nonspherical shape (Matthews, 2003). Besides, surface instability may induce the spheroidization of core-shell nanowires (Kolb et al., 2005; Duan et al., 2008; Schmidt et al., 2008), the buckling of core-shell spheres and cylinders (Tsapis et al., 2005; Cao et al., 2008; Yin et al., 2009a, b), and some other surface morphologies.

In addition to the surface instability in response to external loads, a soft thin film on a substrate may lose its stability spontaneously (Xie et al., 1998). The spontaneous instability causes surface roughness and, therefore, poses a limitation to some industrial applications of thin films (Huang et al., 2008). On the other hand, it can also lead to the formation of various regular patterns with a characteristic wavelength of microns or even nanometers, which are difficult to be produced by using conventional surface fabrication techniques. A deep understanding of the physical mechanisms and critical conditions of spontaneous instability of thin soft films is of paramount significance for the optimal design, manufacture, and manipulation of relevant microscaled/nanosized devices and systems. Therefore, the present paper aims to theoretically investigate the spontaneous surface instability and pattern formation of soft elastic thin films engendered by van der Waals interaction forces. For instance, the spherical and cylindrical configurations with either positive or negative curvatures are considered in detail. Adopting the linear stability analysis method, the critical conditions of the three-dimensional morphological instability for four representative geometric configurations are derived analytically. The effects of substrate curvature, surface energy, thin film thickness, and Poisson's ratio on the surface instability and pattern characteristics are examined.

## 2. Analysis method

### 2.1. Model

Consider a soft thin film with an initially uniform thickness,  $h$ , ideally bonded to a rigid support, as shown in Fig. 1. For simplicity, the film is assumed to be isotropic and linearly elastic.

For a soft film with thickness on the order of micrometers or nanometers, long-range surface/interface interactions associated with electrostatic or van der Waals forces often play a considerable or even dominant role in its deformation, and the effect of surface energy should also be taken into account. Thus, the total free energy of the system,  $\Pi$ , consists of the

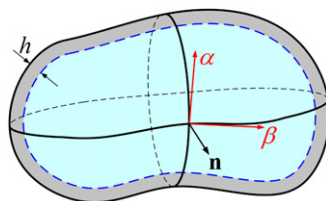


Fig. 1. Schematic of a soft thin film coated on a curved substrate.

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