Surface Instability and Pattern Formation in Two Interacting Incompressible Elastic Films Bonded to Rigid Substrates

ARTICLE *in* LANGMUIR · MARCH 2002

Impact Factor: 4.46 · DOI: 10.1021/la015552p

2 AUTHORS, INCLUDING:

Ashutosh Sharma IITK
Indian Institute of Technology Kanpur
334 PUBLICATIONS **7,407** CITATIONS

Surface Instability and Pattern Formation in Two Interacting Incompressible Elastic Films Bonded to Rigid Substrates

Vijay Shenoy*,† and Ashutosh Sharma‡

Departments of Mechanical and Chemical Engineering, Indian Institute of Technology Kanpur, UP 208 016, India

Received August 24, 2001. In Final Form: December 15, 2001

The surface stability of two interacting (for example, by van der Waals or electrostatic forces) incompressible thin elastic films, one bonded to a substrate and the other to a contactor, is studied, extending the work of Shenoy and Sharma (*Phys. Rev. Lett.* **2001**, *18*, 119−122), who studied the interaction of a film with a rigid contactor. The surfaces of the films roughen spontaneously when the separation is below a critical value. The linear stability analysis indicates that the wavelength of the instability depends strongly on the shear moduli and thicknesses of the films but not on the nature and magnitude of the interaction. Three cases are considered. First, when the films have equal shear moduli but different thicknesses, the wavelength of the instability has an intermediate value between the wavelengths of the instabilities had each of the films been interacting with rigid contactors. Second, if the films have different shear moduli but equal thicknesses, then the wavelength of the instability (approximately equal to thrice the thickness of the film) is identical to that had the films been interacting with rigid contactors. Third, in the more general case when the two films have different shear moduli and thicknesses, the behavior of the critical wavelength is more complex with several interesting features. For example, when the shear moduli of the films are very different, the critical wavelength depends discontinuously on the ratio of thicknesses of the films. The results of this paper are of interest in understanding contact at soft interfaces, peeling of adhesives, cavitation, and so forth and can stimulate further experiments.

1. Introduction

The mechanics of adhesion and contact between two elastic bodies has attracted much attention over the years. Following the classical work of Hertz, an important step in this area was taken by Johnson, Kendall, and Roberts[1] who identified the importance of the interactions between the contacting surfaces. These interactions can be due to the van der Waals force, electrostatic force, and so forth between the contacting surfaces. Indeed, measurements of van der Waals forces between surfaces[2] were available when the now famous Johnson−Kendall−Roberts (JKR) theory of contact[1] was formulated.

More recently, two groups[3,4] have reported experiments aimed at understanding microcontact and adhesion mechanics between two elastic bodies with initial planar topology. The experiments reported in ref 3 used a configuration where a curved glass plate was brought in contact with an elastomeric film between two spacer bars creating a small gap between the film and the glass plate. The authors in ref 4 performed experiments that involved the contact of a flat glass plate with an elastomeric thin film bonded rigidly to a glass substrate. In both of these experimental works, it was observed that the film surface lost planarity when the contacting glass plate reached contact proximity (10−50 nm), resulting in a pattern with a well-defined wavelength. The two key features observed in both sets of experiments are that (a) the wavelength of the pattern depends linearly on the thickness of the film and (b) the wavelength is not affected by the magnitude and nature of the interactions. Similar observations are made by Shull et al.[5] using a spherical contactor.

The theoretical interpretation of this novel elastic instability was first reported by Shenoy and Sharma[6] who argued that the instability occurs due to a competition between the interaction energy of the film with the contactor and the elastic energy due to inhomogeneous deformation in the film. The instability sets in when the ratio of the "stiffness of interaction" and the elastic stiffness (defined as shear modulus divided by the thickness) exceeds a critical value. In addition, they pointed out that the wavelength of the instability is determined solely by the elastic energy of the film and explained the linear dependence of the wavelength on the thickness of the film. A detailed account of this work can be found in ref 7. The origins and nature of this instability are different from those known to exist in solid films[8−11] and fluid films.[12−15]

The purpose of this paper is to extend the work of ref 6 to the case when the contactor also has a film bonded to it as shown in Figure 1. The contacting film may have different properties in that the shear modulus and

* Corresponding author. Tel: +91-512-597307. Fax: +91-512-597408. E-mail: vbshenoy@iitk.ac.in.

† Department of Mechanical Engineering.

‡ Department of Chemical Engineering.

(1) Johnson, K. L.; Kendall, K.; Roberts, A. D. *Proc. R. Soc. London* **1971**, *A324*, 301−313.

(2) Tabor, D.; Winterton, R. H. S. *Proc. R. Soc. London* **1969**, *A312*, 435−450.

(3) Ghatak, A.; Chaudhury, M. K.; Shenoy, V.; Sharma, A. *Phys. Rev. Lett.* **2000**, *85*, 4329−4332.

(4) Mönch, W.; Herminghaus, S. *Europhys. Lett.* **2001**, *53*, 525−531.

(5) Shull, K. R.; Flanigan, C. M.; Crosby, A. J. *Phys. Rev. Lett.* **2000**, *84*, 3057−3060.

(6) Shenoy, V.; Sharma, A. *Phys. Rev. Lett.* **2001**, *86*, 119−122.

(7) Shenoy, V.; Sharma, A. *J. Mech. Phys. Solids*, in press.

(8) Asaro, R. J.; Tiller, W. A. *Metall. Trans.* **1972**, *3*, 1789−1796.

(9) Grinfeld, M. *J. Nonlinear Sci.* **1993**, *3*, 35−83.

(10) Srolovitz, D. *Acta Metall.* **1989**, *37*, 621−625.

(11) Ramirez, J. C. *Int. J. Solids Struct.* **1989**, *25*, 579−589.

(12) Herminghaus, S.; Jacobs, K.; Mecke, K.; Bischof, J.; Fery, A.; Ibn-Elhaj, M.; Schlagowski, S. *Science* **1998**, *282*, 916−919.

(13) Sharma, A.; Khanna, R. *Phys. Rev. Lett.* **1998**, *81*, 3463−3466.

(14) Reiter, G.; Khanna, R.; Sharma, A. *Phys. Rev. Lett.* **2000**, *84*, 1432−1435.

(15) Schäffer, E.; Thurn-Albrecht, T.; Russel, T. P.; Steiner, U. *Nature* **2000**, *403*, 874−877.

10.1021/la015552p CCC: $22.00
Published on Web 01/26/2002

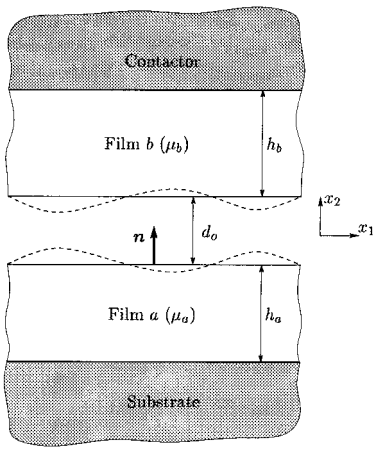

Figure 1. A thin elastic film bonded to a rigid substrate interacting with another film bonded onto a contactor. The dashed lines show possible inhomogeneous deformation of the films when instability sets in. (Distances are not to scale.)

thickness may be different from those of the film bonded to the substrate. It is shown that one of key features, that is, the independence of the wavelength of the instability on the nature of interaction, is unchanged. Although the critical wavelength does not depend on the nature and strength of the interaction, it does depend strongly on the thicknesses and moduli of the films. It is shown that the relative stiffnesses of the films play an important role in the determination of the wavelength of the instability when the films have widely differing properties. The complete dependence of the wavelength on the thicknesses and moduli of the film is obtained. It is hoped that this will greatly aid in the fundamental understanding of friction and adhesion when two soft surfaces are in contact. This paper is also written with a purpose of motivating further experiments in the kinds of systems indicated in Figure 1 with a view toward exploiting them for the creation of mesoscale patterns in solid films for a host of nanotechnology applications. The paper is organized as follows. The next section contains a stability analysis of two interacting films. Results of the analysis are presented and discussed in section 3. The findings of this paper are summarized in section 4. The Appendix at the end of the paper contains a short calculation of the normal traction along the surface of an incompressible film subjected to a sinusoidal deformation.

2. Stability of Two Interacting Films

2.1. Model Description. A schematic depiction of the model considered for the study of stability of interacting thin films is shown in Figure 1. The system consists of a substrate−contactor configuration where both the substrate and the contactor have thin films (possibly of different materials) bonded to them. The substrate film (called film a) has a thickness h_a, while the contactor film (called film b) has a thickness h_b. A coordinate system described by coordinates (x_1, x_2) is used to describe position vectors. The positive 2 direction is the outward normal of film a, while the negative 2 direction points in the normal direction to the surface of the film b. We consider only plane strain deformation of the system.

The equilibrium configuration of this system is determined by the potential energy,

$$\Pi = \int_V W(\epsilon)\, dV - \int_S U((\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n})\, dS \qquad (1)$$

where W is the strain energy density, ϵ is the strain tensor, $\mathbf{u}$ is the displacement vector (the superscript a denotes value in film a, etc.) with V being an appropriate measure of the volume (includes volumes of both the films), and S is the interfacial area of the two films over which they interact. The function U represents the interaction potential between the two films; it is this term that gives rise to interesting physics in this system. We make two key physical assumptions in writing the total potential energy: (i) the contribution from the surface energies of the films is negligible and (ii) the films are considered to be made of incompressible elastic materials. Both of these assumptions are valid in physical systems where such instabilities are triggered, as shown earlier.[6]

The attractive interaction between the surfaces results from van der Waals force, electrostatic force, and so forth. The potential U therefore denotes a generic interaction potential. If the potential is due to the attractive van der Waals interaction, then U is described by

$$U((\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n}) = \frac{1}{12\pi}\frac{A}{(d_0 - (\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n})^2} \qquad (2)$$

where A is the Hamaker constant (of the order of 10^{-19} J), and d_0 is the initial constant distance between the surfaces of the two films, that is, the gap thickness. The strength of the interaction potential is determined by the local gap thickness $(d_0 - (\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n})$. The contactor is imagined to be brought in the proximity of the substrate by reducing the distance d_0, the interesting quantity being the distance d_c at which the attractive interactions are strong enough to trigger the surface instability and roughening in the system.

Linear stability analysis is performed using linear kinematics and a linearized interaction potential. To this end, the interaction potential is expanded in a power series about the reference state of the undeformed films and terms of up to quadratic order in $(\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n}$ are retained,

$$U((\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n}) \approx U_0 + F(\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n} + \frac{1}{2}Y((\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n})^2 \qquad (3)$$

where

$$U_0 = U(0) \qquad F = U'(0) \qquad Y = U''(0) \qquad (4)$$

The quantity Y, called the *interaction stiffness*, is of importance and governs the stability of the system. One of the aims of the analysis is to find the condition(s) on Y under which instability sets in. It is to be noted that the expansion in eq 3 is valid for any kind of interaction (be it van der Waals, electrostatic, etc.) that depends smoothly on the gap between the films. Indeed, this is not a limitation since almost all physical interactions of interest depend smoothly on the gap thickness.

The above linearization gives an expression for the potential energy as

$$\Pi_a = \int_V W(\epsilon)\, dV - \int_S \left(U_0 + F(\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n} + \frac{1}{2}Y((\mathbf{u}^a - \mathbf{u}^b)\cdot\mathbf{n})^2\right) dS \qquad (5)$$

$$W(\epsilon) = \begin{cases} \frac{\mu_a}{2}\epsilon : \epsilon & \text{in film a} \\ \frac{\mu_b}{2}\epsilon : \epsilon & \text{in film b} \end{cases} \qquad (6)$$

where μ_a and μ_b are the shear moduli of the films a and b, respectively. The equilibrium displacement fields in the films minimize the potential energy (eq 5) while satisfying the rigid boundary conditions at the film−substrate interface in film a and the film contactor interface in film b. In addition, the stresses derived from these displacement fields satisfy the condition of vanishing shear stress σ_{12} at the surface in both the films. The normal stresses satisfy the condition

$$\sigma_{22}^{a} = F + Y(u_2^{a} - u_2^{b}) \tag{7}$$

$$\sigma_{22}^{b} = F + Y(u_2^{a} - u_2^{b}) \tag{8}$$

at their respective surfaces.

Homogeneous Solution. Since both films are incompressible, the homogeneous solution has displacements vanishing everywhere in both films, and the stress state in both films is one of constant pressure (equal to F). The point in question is the stability of this homogeneous solution; that is, for what value(s) of Y does instability occur?

2.2. Stability Analysis. To study the stability of the system above, the homogeneous solution is perturbed by bifurcation fields (denoted by u_i here and henceforth) such that the surfaces of films a and b have displacements of

$$u_2^{a}(x_1) = \alpha \cos(kx_1) \tag{9}$$

and

$$u_2^{b}(x_1) = \beta \cos(kx_1) \tag{10}$$

respectively. The *additional stresses* produced by these fields satisfy

$$\sigma_{12}^{a} = 0 \qquad \sigma_{12}^{b} = 0 \tag{11}$$

and

$$\sigma_{22}^{a} = Y(u_2^{a} - u_2^{b}) \tag{12}$$

$$\sigma_{22}^{b} = Y(u_2^{a} - u_2^{b}) \tag{13}$$

along the interacting surfaces of the films. All symbols have obvious meanings.

It is shown in the Appendix that the stress σ_{22}^{a} at the surface of a film a whose displacement is $u_2^{a}(x_1) = \alpha \cos(kx_1)$ with $\sigma_{12}^{a} = 0$ is given by

$$\sigma_{22}^{a}(x_1) = 2\mu^{a}\, S(h_a k)\, k\alpha \cos(kx_1) \tag{14}$$

and the stress $\sigma_{22}^{b}(x_1)$ along the surface of the film b is

$$\sigma_{22}^{b}(x_1) = -2\mu^{b}\, S(h_b k)\, k\beta \cos(kx_1) \tag{15}$$

where S is a nondimensional function defined as

$$S(\xi) = \frac{1 + \cosh(2\xi) + 2\xi^2}{\sinh(2\xi) - 2\xi} \tag{16}$$

For future discussion, we note that $S(\xi) \to 1/\xi^3$ as $\xi \to 0$, and $S(\xi) \to \coth(\xi)$ when $\xi \to \infty$.

Substitution of eqs 14 and 15 in eqs 12 and 13 leads to the following homogeneous set of equations for α and β:

$$\begin{pmatrix} 2\mu_a k S(h_a k) - Y & Y \\ -Y & Y - 2\mu_b k S(h_b k) \end{pmatrix} \begin{pmatrix} \alpha \\ \beta \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \tag{17}$$

The condition for the existence of nontrivial bifurcation fields, that is, the condition for the existence of nontrivial solutions for α and β, is obtained by setting the determinant of the coefficient matrix to zero resulting in a relation between the interaction stiffness Y and the wavenumber k of the bifurcation field,

$$Y = \frac{2k\mu_a\mu_b\, S(h_a k)\, S(h_b k)}{\mu_a\, S(h_a k) + \mu_b\, S(h_b k)} \tag{18}$$

If, for a given value of Y, there is at least one real value of k that solves eq 18, then the homogeneous solution is unstable and the films deform inhomogeneously. The *lowest value* of Y for which bifurcations are possible is called the *critical interaction stiffness* denoted by Y_c. The wavenumber(s) of the mode(s) that satisfies (satisfy) eq 18 for $Y = Y_c$ is (are) called the critical mode(s) and the (these) wavenumber(s) is (are) denoted by k_c.

At this point, there are four parameters μ_a, μ_b, h_a, and h_b that enter into the determination of the stability of the system. The discussion to follow is much simplified on introduction of effective parameters; to this end, we define

$$\mu = \frac{\mu_a\mu_b}{\mu_a + \mu_b} \tag{19}$$

and

$$h = h_a + h_b \tag{20}$$

We also introduce nondimensional parameters M and H defined as

$$\frac{\mu}{\mu_a} = M \qquad \frac{\mu}{\mu_b} = (1 - M) \tag{21}$$

and

$$h_a = Hh \qquad h_b = (1 - H)h \tag{22}$$

When $\mu_a \ll \mu_b$, $M \to 1$, and $M \to 0$ if $\mu_b \ll \mu_a$. Similarly, when $h_a \gg h_b$, $H \to 1$, while $H \to 0$ implies $h_b \gg h_a$. Another important quantity of interest is the effective elastic stiffness K_{eff} of the two-film system defined as

$$K_{eff} = \frac{\dfrac{\mu_a}{h_a}\dfrac{\mu_b}{h_b}}{\dfrac{\mu_a}{h_a} + \dfrac{\mu_b}{h_b}} = \frac{\mu_a\mu_b}{\mu_a h_b + \mu_b h_a} = \frac{\mu}{h}\frac{1}{((1 - H)(1 - M) + HM)} \tag{23}$$

Based on the above definitions, the expression for the interaction stiffness in eq 18 can be recast in the following nondimensional form:

$$\frac{Y}{K_{eff}} = \frac{2((1 - H)(1 - M) + HM)q\, S(Hq)\, S((1 - H)q)}{(1 - M)\, S(Hq) + M\, S((1 - H)q)} \tag{24}$$

where $q = hk$.

The results for the special case of a rigid contactor, that is, when $\mu_b \gg \mu_a$, obtained by Shenoy and Sharma[6] can be

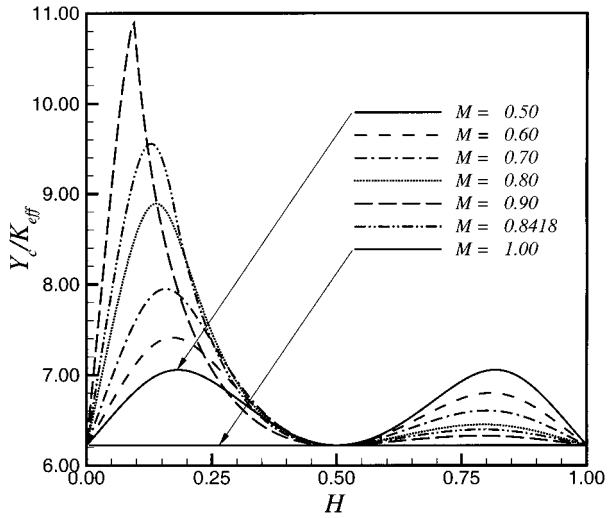

Figure 2. The critical interaction stiffness as a function of H for various values of M.

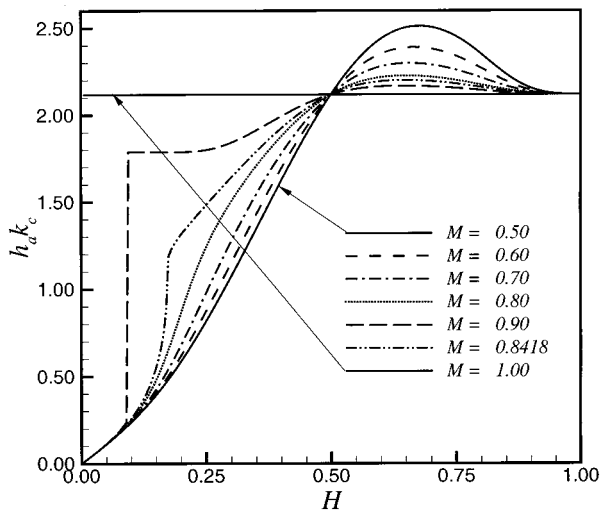

Figure 3. The wavelength of the critical mode as a function of H for various values of M.

immediately recovered. Thus, consider the case when $M \rightarrow 1$, $K_e \rightarrow \mu_a/h_a$, and

$$\frac{h_a Y}{\mu_a} = 2Hq\ S(Hq) = 2h_a k\ S(h_a k) \qquad (25)$$

Under the above conditions, eq 25 gives the critical interaction stiffness as $Y_c = 6.22\mu_a/h_a$ and $h_a k_c = 2.12$, which is precisely the result in ref 6. Another route to the same result is to let $h_a \gg h_b$, that is, as $H \rightarrow 1$. In this case, $K_e \rightarrow \mu_a/h_a$ again, and

$$\frac{h_a Y}{\mu_a} = 2Hq\ S(Hq) = 2h_a k\ S(h_a k) \qquad (26)$$

where the property of the function S near 0, noted above, is used. It can be easily shown that similar results are obtained when either $\mu_b \ll \mu_a$ ($M \rightarrow 0$) or $h_b \gg h_a$ ($H \rightarrow 0$).

In this section, we have derived the condition for the onset of instability in a system with two interacting incompressible elastic films. In addition, it is shown that the results of Shenoy and Sharma[6] for the case of a film interacting with a rigid contactor are special cases of the present formulation. The more general case is taken up for discussion in the next section.

3. Results and Discussion

The nondimensional quantities M and H introduced in the last section can take any value from 0 to 1. However, due to the symmetry of eq 24, it is evident that the results for a given value of M and H are physically identical to that of $(1 - M)$ and $(1 - H)$. Due to this reason, only the regime $^1/_2 \le M \le 1$ and $0 \le H \le 1$ is considered. Results are discussed in three categories: (i) $\mu_a = \mu_b$, $h_a \neq h_b$; (ii) $\mu_a \neq \mu_b$, $h_a = h_b$; (iii) $\mu_a \neq \mu_b$, $h_a \neq h_b$. The results of all these cases are plotted in two sets of graphs shown in Figures 2 and 3.

Just as in the case of a film interacting with a rigid contactor, the wavelength of the instability does not depend on the magnitude and nature of the interaction forces but depends only on the thicknesses of the films and their moduli, that is, on the parameters H and M. Of course, a critical interaction stiffness Y_c which governs the strength of the interaction force is needed for the onset of roughening. Therefore, the critical distance at which the instability sets in is different for different types of interactions.

3.1. Films with Equal Shear Moduli. When the shear moduli of the two films are equal ($\mu_a = \mu_b$), $M = ^1/_2$, $\mu = \mu_a/2$ and $K_{eff} = H\mu_a/h_a$. The results for Y_c and k_c correspond to $M = 0.5$ in Figures 2 and 3. The magnitude of nondimensional critical interaction stiffness Y_c/K_{eff} increases with decreasing H (starting from $H = 1$), then attains a peak, and falls thereafter as H approaches $^1/_2$. If h_a is kept fixed and h_b is changed to change H, then increasing Y_c/K_{eff} does not imply an increase in Y_c, since K_{eff} falls linearly with H. Thus, for a fixed h_a, the effective stiffness of the system reduces with decreasing H. For example, when $H = ^3/_4$, that is, $h_b/h_a = ^1/_3$, the value of $K_e = 3\mu_a/4h_a$ and the value of $Y_c/K_e = 6.92$ or $Y_c = 5.19\mu_a/h_a$ which is *less* than $Y_c = 6.22\mu_a/h_a$ when film a interacts with a rigid contactor. Looking at the same example from the point of view of film b, we note that $K_e = (1 - H)\mu_b/h_b$ and $Y_c = 1.73\mu_b/h_b$ which is again less than the value $6.22\mu_b/h_b$. In fact, when films have equal thicknesses and equal shear moduli, the value of $Y_c = 3.11\mu_a/h_a$. The conclusion of these observations is that the magnitude of the critical interaction stiffness required for instability is less than that when either of the films is interacting with a rigid contactor. Physically, this can be understood by the fact that introduction of a second elastic film on the contactor makes the system *more compliant*; that is, the effective elastic stiffness of the system comes down. Thus, the elastic energy penalty required to cause an inhomogeneous deformation in the system also comes down. As a consequence, the absolute value of the interaction stiffness required to trigger the instability is reduced. Moreover, the actual value of Y_c is governed by the more compliant film.

Turning now to a discussion of the wavenumber of the critical mode, we note that as H decreases from 1, $h_a k_c$ increases and then falls subsequently. For values of H between $^1/_2$ and 1, the instability triggers a shorter wavelength mode in film a as compared to when film a interacts with a rigid contactor. For example, when $H = ^3/_4$, the value of $h_a k_c = 2.45$; in terms of film b, the value of k_c is $h_b k_c = 0.82$, that is, the wavelength of the critical mode is much larger than that if film b had been interacting with a rigid contactor. The key point is that the wavelength of the instability in this regime is *intermediate* to critical wavelengths determined by the thicknesses of the participating films. As the thicknesses are made equal, that

is, when $H = {}^1/_2$, the critical wavelength becomes $h_a k_c = 2.12$ which is exactly the wavelength had either of the films been interacting with a rigid contactor. When $M = {}^1/_2$, the case of $H < {}^1/_2$ is physically identical to the case $H > {}^1/_2$. These results can be interpreted physically as follows. The dependence of elastic energy in the film on the wavenumber k is determined solely by the thickness of the film, or more precisely by the function S (defined in eq 16) which depends on the thickness of the film; the shear modulus is a mere multiplicative parameter. The elastic energy (per unit length of the film) is a minimum when the wavenumber is equal to 2.12/(thickness). However, when there are two films with *very different thicknesses*, the total elastic energy in the system (sum of elastic energies in the two films) can be minimized by choosing an intermediate wavelength.

3.2. Films with Equal Thicknesses. When the films are made of equal thickness $h_a = h_b$, the parameter $H = {}^1/_2$. For this case, the effective stiffness $K_e = \mu/h_a$, and the nondimensional critical interaction stiffness is independent of the value of M, with $Y_c = 6.22\mu/h_a$. Since $\mu < \mu_a$ and $\mu < \mu_b$, the actual magnitude of the critical interaction stiffness is less than the critical interaction stiffness of the films if they had been interacting with rigid contactors. This can, again, be understood by the argument that the effective stiffness of the system comes down on introduction of the second film on the contactor. The critical wavenumber, however, does not depend on M as is evident from Figure 3 ($H = {}^1/_2$). The critical wavelength is *equal* to the wavelength had the films been interacting with rigid contactors. Again, this can be understood from the argument stated above that the elastic energy of a mode is determined by the thickness of the film via the function S and the moduli are multiplicative constants in the energy expression. This idea is evident from eq 24 on substitution of $H = {}^1/_2$.

3.3. Films with Unequal Thicknesses and Moduli. The physics of surface instability in the general case can be understood based on the results of the previous two cases. When $M > {}^1/_2$, film a is softer than film b, that is, $\mu_a < \mu_b$. When M is close to ${}^1/_2$, the qualitative features of the instability are unchanged. The nondimensional interaction stiffness Y_c/K_{eff} increases with decreasing H from 1 and falls as H approaches ${}^1/_2$. On further decrease of H, Y_c/K_{eff} increases and decreases again to 6.22 as H approaches 0. The behavior of the critical wavelength is thus qualitatively identical to that of the case when $M = {}^1/_2$.

As M is made close to 1 ($M < 1$), a qualitative change appears in the solution. In fact, when $M = 0.8418$ (corresponding to $\mu_b/\mu_a = 5.32$), the critical wavenumber changes suddenly as a function of H at about $H = 0.1756$ (corresponding to $h_b/h_a = 4.69$). For larger values of M, as H is reduced, the critical wavenumber remains a constant (and close to the wavenumber $h_a k_c = 2.12$) but jumps suddenly to a much lower value; the value of H at which the jump occurs is called H_J. This is evident when $M = 0.9$ (shown in Figure 3) where the wavenumber of the critical mode $h_a k_c \approx 1.79$ when $H < 0.25$; however, at $H = 0.095 = H_J$ (corresponding to $h_b/h_a = 9.52$), the critical wavelength becomes $h_a k_c = 0.22 \rightarrow h_b k_c = 2.1$, that is, the wavenumber of the instability changes to the critical wavenumber of film b had it been interacting with a rigid contactor. The value of H_J, that is, the nondimensional value of H at which the critical wavenumber jumps from a higher value k_h to a lower value k_l, depends on the ratio of the shear moduli; this dependence is plotted in Figure 4. It is evident that H_J decreases with M, as is expected from the physical arguments presented below. The two

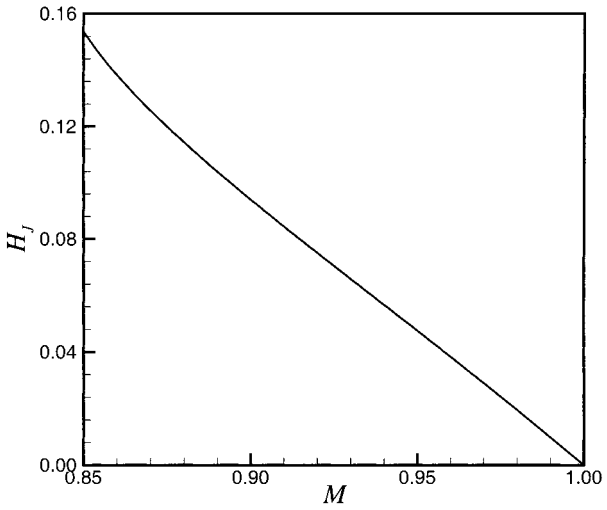

Figure 4. The nondimensional parameter H_J (that characterizes the difference in the thicknesses of the films) at which the critical wavenumber jumps, as a function of M.

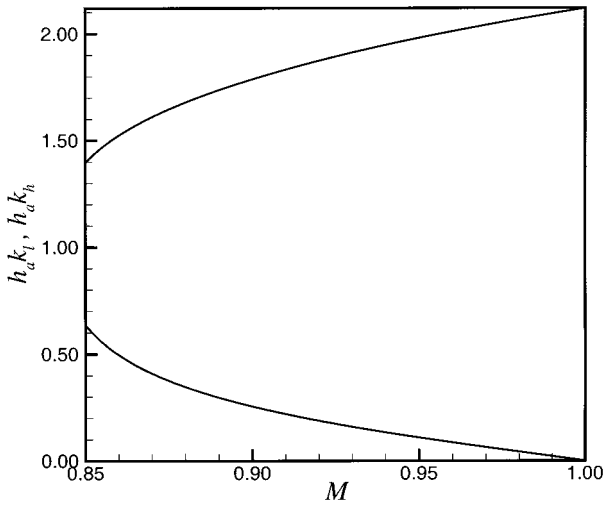

Figure 5. The lower ($h_a k_l$) and higher ($h_a k_h$) wavenumbers at $H = H_J$, as a function of M.

wavenumbers at H_J are plotted in Figure 5. Again, as M approaches 1, $h_a k_h$ approaches 2.12, the value determined by the thickness of film a, and $h_a k_l$ approaches 0 (it can, in fact, be shown that $h_b k_l$ approaches 2.12).

From a physical perspective, the total elastic energy is the sum of elastic energies in the films which are products of their respective shear moduli with the S function defined in eq 16. When the elastic moduli are very different, the total energy of the system can have multiple minima, which implies that Y as a function of q defined in eq 24 has multiple minima. Indeed, a plot of Y as a function of $h_a k$ shown in Figure 6 clearly shows the presence of multiple minima; additionally, the value of $h_a k$ where the minimum is attained changes to a smaller value of $h_a k$ as H is reduced. The key physical idea is that instability is governed by the film that has a smaller stiffness (the compliant film). Although μ_b is much larger than μ_a, h_b can be made larger than h_a to the extent that the stiffness of film b is smaller than that of film a. Thus, when H is lowered, the critical wavenumber of the instability of the system jumps from a wavenumber close to the critical wavenumber determined by film a (as if interacting with a rigid contactor) to that of film b (as if interacting with a rigid substrate).

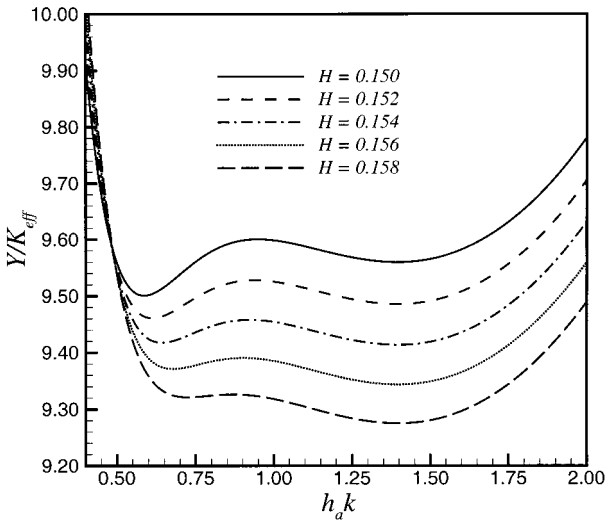

Figure 6. The interaction stiffness Y/K_{eff} as a function of $h_a k$ showing multiple minima for different values of H with $M = 0.85$. Note how the value of $h_a k$ at which Y/K_{eff} attains a minimum changes as H is reduced.

4. Summary and Conclusion

This paper extends the work of Shenoy and Sharma,[6] who studied the interaction of a soft thin film with a rigid contactor, to the case where the contactor also has a soft film bonded to it. This new analysis uncovers some interesting new results that are summarized below.

1. Surface instability and roughening set in when the attractive intersurface force manifested in the interaction stiffness exceeds a critical value which is determined by the shear moduli and thicknesses of the interacting films. Thus, the instability occurs below a critical intersurface distance, which depends on the nature of interactions. The magnitude of the critical interaction stiffness is smaller than that of the critical interaction stiffnesses of the individual films had they been interacting with rigid contactors. Thus, it is "easier" for the instability to occur in the two-film system.

2. The critical wavelength is determined only by the thicknesses and shear moduli of the films and does not depend on the nature and magnitude of the interaction (i.e., the critical wavelength does not depend on the critical interaction stiffness). This may be contrasted with liquid films (see, for example, ref 16) where a linear stability analysis indicates that the wavelength of stability depends strongly on the interaction. While the linear stability analysis does predict the wavelength of the instability, a fully nonlinear analysis is required to determine the amplitude of roughness that sets in. This nonlinear analysis will be taken up for study in future publications.

3. When the films have equal shear moduli, the critical wavenumber of the instability depends on the relative thicknesses of the films. When the thicknesses of the films are very different, the wavelength of the instability is intermediate between the wavelength of the instability in each of the films had they been interacting with rigid contactors. However, when the thicknesses of the films are equal, the wavelength of the instability is identical to the case when the films interact with rigid contactors.

4. When the films have equal thicknesses but different shear moduli, the magnitude of the critical interaction stiffness is less than that of each of the films had they been interacting with rigid contactors. The wavelength of the critical mode, however, is equal to that when the films interact with rigid contactors and is independent of the shear moduli of the films.

5. When the films are of both unequal thicknesses and moduli, the behavior is dominated by the less stiff films (stiffness being the ratio of the shear modulus to the thickness of the film). As long as the modulus of the contacting film is less than about 5 times that of the substrate film, the behavior of the solution is qualitatively the same as that discussed in the above cases. However, when the modulus of the contacting film is larger than 5.32 times the thickness of the film on the substrate, the wavelength of the instability depends strongly on the thicknesses of the films. As the thickness of the contacting film is increased, the wavelength of the instability jumps from a value determined by the thickness of the substrate film to that determined by the contacting film.

It is hoped that the paper will further stimulate experiments along the lines of refs 3−5 to verify the results presented here. The analysis presented here neglects the effect of surface energies and the compressibility of the films. The inclusion of these effects along with the viscosity of the films will be presented elsewhere.[17]

Acknowledgment. V.S. wishes to thank DST, India, for support of this work under the Fast Track Scheme.

Appendix: Stresses along the Surface of an Incompressible Film Bonded to a Substrate with Sinusoidal Surface Deformation

The aim of this Appendix is to outline the determination of the normal traction along the surface of an incompressible film bonded to a rigid substrate, the surface of which is deformed sinusoidally. The main result of this Appendix is the formula used in eqs 14 and 15.

Consider an incompressible elastic film described by coordinates (x_1, x_2) of thickness h bonded to a rigid substrate such that the free surface of the film has coordinate $x_2 = 0$ and the interface between the film and the substrate has coordinate $x_2 = -h$. The film has a shear modulus μ. The boundary value problem has the following boundary conditions:

$$u_1(x_1, -h) = u_2(x_1, -h) = 0 \tag{27}$$

$$u_2(x_1, 0) = \alpha \cos(kx_1) \qquad \sigma_{12}(x_1, 0) = 0 \tag{28}$$

The equilibrium equation in terms of the displacements is

$$\mu\, u_{i,jj} + p_{,i} = 0 \tag{29}$$

where p is the pressure field. The incompressibility condition is expressed as

$$u_{i,i} = 0 \tag{30}$$

A general solution of the set of differential equations that anticipates the boundary conditions (27) and (28) is

$$u_1(x_1, x_2) = -\frac{\alpha}{k}((B + k(A + Bx_2))e^{kx_2} + (D - k(C + Dx_2))e^{-kx_2}) \sin(kx_1) \tag{31}$$

$$u_2(x_1, x_2) = \alpha((A + Bx_2)e^{kx_2} + (C + Dx_2)e^{-kx_2}) \cos(kx_1) \tag{32}$$

(16) Sharma, A. *Langmuir* **1993**, *9*, 861−869.
(17) Sarkar, J.; Shenoy, V.; Sharma, A. Manuscript under preparation.

$$p(x_1, x_2) = -2\mu\alpha(Be^{kx_2} + De^{-kx_2})\cos(kx_1) \quad (33)$$

The constants A, B, C, and D can be determined from the boundary conditions (27) and (28). The solution is

$$A = \frac{1 + e^{2kh} - 2kh(1 - kh)}{2\sinh(2kh) - 4kh} \qquad C = 1 - A \quad (34)$$

$$B = -\frac{k(1 + e^{2kh} - 2kh)}{2\sinh(2kh) - 4kh} \qquad D = k + B \quad (35)$$

From these relations, the expression for the normal component of traction along the surface of the film can be derived as

$$\sigma_{22}(x_1, 0) = 2\mu\, u_{2,2} + p = 2\mu(2A - 1)ka\cos(kx_1) \quad (36)$$

On substitution of the solution for A from eq 34, the expression simplifies to

$$\sigma_{22}(x_1, 0) = 2\mu\, S(hk)\, k\alpha\cos(kx_1) \quad (37)$$

where the function S is defined in eq 16.

LA015552P