THE MORPHOLOGY AND FOLDING PATTERNS OF BUCKLING-DRIVEN THIN-FILM BLISTERS

MICHAEL ORTIZ and GUSTAVO GIOIA
Division of Engineering, Brown University, Providence RI 02912, U.S.A.

(Received 8 June 1993)

ABSTRACT

Thin films and coatings in a state of residual compression can, under appropriate conditions, decohere and buckle away from the substrate to form blisters. These blisters are often observed to adopt intricate shapes and to fold into complex patterns. In this paper, such shapes and patterns are given an energetic interpretation, i.e. they follow as energy minimizers. We formulate the energy of the film by recourse to von Kármán theory of moderate deflections of a plate. The energy functional has the following key properties: it contains two terms, namely, the membrane and bending energies, the latter being a singular perturbation of the former; and the membrane energy functional is nonconvex and, consequently, its infimum is generally not attained. In keeping with the conventional mathematical treatment of these problems, we construct solutions by a matched asymptotic expansion. The outer solution follows by membrane energy minimization and determines the essential folding pattern of the film. The inner solution is obtained by fitting boundary layers at sharp edges in the membrane solution. The film deflections thus constructed are found to match, in surprising detail, the observed complex folding patterns adopted by delaminated films. In addition, the boundary layer analysis permits one to accord a well-defined line tension to sharp edges in the membrane solution, and, in particular, to the boundary of the blister. This provides a simple device for assessing the configurational stability of some blister morphologies. In particular, the analysis predicts the transition from straight-sided to telephone-cord morphologies at a critical mismatch strain.

1. INTRODUCTION

The mechanical failure of thin films subjected to biaxial compression often occurs by delamination of the film from pre-existing interfacial flaws. The compressed film buckles away from the substrate, which induces further delamination and, under appropriate conditions, may lead to the catastrophic failure of the component. Initially, small flaws tend to have smooth, nearly circular, boundaries and to exhibit smooth deflections. As the flaw grows in size, the film is observed to fold and the boundary to become wavy (ARGON et al., 1980a, b; HUTCHINSON et al., 1992). A relatively common occurrence is for the flaw to issue eventually one or more telephone-cord blisters which grow at their tips (HUTCHINSON et al., 1992; THOULESS, 1993). Straight-sided blisters are sometimes also observed (THOULESS, 1993). Blisters may also grow in an equiaxed fashion up to large sizes, in which case the film is observed to adopt intricate folding patterns accompanied by fine waviness of the boundary (ARGON et al., 1989a, b).
The delamination of thin films has been investigated by a number of researchers [see the reviews of Alexopoulos and O'Sullivan (1990) and Hutchinson and Suo (1991)]. The studies to date have been based on conventional elasticity theory and interfacial fracture mechanics, and have primarily been concerned with the stability of blisters of simple shapes, such as circular and straight-sided flaws. However, conventional methods of analysis tend to become unwieldy when applied to arbitrary domains or to very large blisters, in which intricate folding patterns may develop. As a consequence, the folding patterns of large blisters and some of the commonly observed boundary morphologies have defied effective analytical treatment.

In this paper we pursue a different line of inquiry based on energy methods. We begin by noting that, in the limit of thin films, the bending energy of the film constitutes a singular perturbation superposed on the membrane energy. This suggests a solution of the problem by matched asymptotic expansions: the outer solution minimizes the membrane energy of the film, and the inner solution is obtained by fitting boundary layers to the sharp edges which are present in the membrane solution. In pursuing this program, however, an essential difficulty is the lack of convexity of the membrane energy when regarded as a functional of the film deflections. In other fields of application, such as phase transitions and micromagnetics, this lack of convexity is tied to the emergence of fine microstructures, such as twinning or magnetic domains (James and Kinderlehrer, 1990, 1993; Kohn and Müller, 1992).

In predicting these microstructures, the so-called direct methods of the calculus of variations have proven particularly effective. These methods seek to characterize solutions directly as energy minimizers, instead of as solutions of the Euler equations of the energy functional. In this context, microstructures naturally arise as a byproduct of energy minimization. Similarly, the intricate folding patterns exhibited by large blisters can be accorded an energetic interpretation as energy minimizers. In Section 4, we demonstrate by way of example how the membrane energy can indeed be fully relaxed by appropriately folding the film. Alternative constructions have been considered by Pipkin (1986a, b) as a basis for deriving tension field theories (Reissner, 1938).

Unfortunately, membrane energy minimization fails to produce a unique folding pattern unless subsidiary conditions are formulated. This difficulty is endemic in problems involving singularly perturbed nonconvex functionals, and has been addressed in the mathematical literature (Modica, 1987; Sternberg, 1988). A method which originated with the pioneering work of de Giorgi (De Giorgi and Franzoni, 1975) is to resort to the singular term in the energy functional for selecting a preferred outer solution. In the present context, the idea is to select the membrane solution containing the least possible bending energy. Because membrane solutions can exhibit sharp edges at which the bending energy density is undefined, the measure of bending energy used to select the preferred membrane solution has to be defined with some care. An approximate definition can be derived simply by a local boundary layer analysis which we adapt from the work of Modica (1987) and Sternberg (1988). By virtue of this analysis, sharp edges can be accorded a well-defined energy per unit length, or line tension. The problem is then to construct a membrane solution having the least possible edge and boundary energy.

We conjecture that a characterization of the solution can be achieved by identifying various possible solutions of all planar blisters and then comparing the results of subsequent studies (caustics in a simple model for the membrane) with the considerations of the analysis of the analytic solutions (1989a, b).

The fold patterns of blisters can involve intricate branching and refinement of the boundary layer. The resulting sharp edges exhibit a detailed relationship between fold patterns and energetics which may depend on the relative contributions of energy balance of the blister layers. Beyond this, the intrinsic problem of delaminating thin films is one which is consistent with a transition in the behavior of the film, with a transition in the strain field, and with a transition in the curvature of the film.

The paper is organized in Sec 5 with Sec 6 being devoted to the character of the membrane energy, and Sec 7 to the two-variant model to relax fulfillment of arbitrary solutions. A separate section which includes other potential and additional evidence is provided in Sec 8. The implications of the delamination of film systems are discussed in Sec 6.
We conjecture that such a preferred membrane solution is the envelope of all cones of a characteristic slope supported in the domain of the blister. The deflections are, therefore, maximal solutions of the Eikonal equation. The preferred membrane solution can also be defined by Nadai's sand-heap construction, or as the envelope of all planes tangent to the boundary having a characteristic slope. Bending can subsequently be taken into account by fitting boundary layers to the sharp edges (caustics in the geometrical optics analogy; ridges in the sand-heap analogy) of the membrane solution. An alternative, albeit approximate, means of taking bending into consideration is by subjecting the membrane solution to Laplacian smoothing. Justification for the proposed construction is found in the excellent agreement between the analytical solutions and the deflection patterns observed by ARGON et al. (1989a, b).

The folding patterns which fully relax the membrane energy of the film invariably involve increasingly finer features. Bending effectively checks this process of indefinite refinement and introduces a lower bound for the spacing of the folds. Indeed, a boundary layer analysis permits the assignment of a well-defined width to the sharp edges exhibited by the membrane solution. This width sets the minimum spacing between folds, and is found to correlate closely with the wavelength of the boundary undulations observed in large blisters. This suggests that such undulations are directly induced by the internal folding of the film. In other cases, the geometry of the boundary may depend critically on the fracture properties of the interface. Indeed, a simple energy balance argument reveals that the fracture toughness of the interface, in conjunction with line tension, determine an upper bound for the radius of curvature of the boundary. In addition, straight boundaries, such as found in straight-sided blisters, are determined to be stable only up to a critical value of the mismatch strain. Beyond this critical point, straight boundaries are predicted to lose stability and to delaminate spontaneously so as to attain the critical curvature. These predictions are consistent with recent observations of Thouless (private communication), who induced a transition from a straight-sided to a telephone-cord morphology in a mica-Al system in the laboratory by cooling down the film, thus presumably increasing the mismatch strain beyond the critical value. These observations additionally suggest that the telephone-cord morphology is but the juxtaposition of arcs of circles at the critical radius of curvature, and that it arises in the supercritical regime as a consequence of the curvature constraint.

The paper is structured as follows. The energy functional of the film is formulated in Section 2 by recourse to von Kármán theory of plates. The perturbative character of bending in the thin-film limit and the lack of convexity of the membrane energy functional are discussed in Section 3. In Section 4 we give a simple two-variant construction for folding a straight-sided semi-infinite blister so as to relax fully its membrane energy. In Section 5 we address the case of blisters of arbitrary shape. In particular, in Section 5.3 we give a simple analytical construction which appears to capture the essential folding patterns observed in large blisters, as demonstrated in Section 5.5 by comparison with the available experimental evidence. The effect of bending is discussed in Sections 5.2 and 5.4. Finally, the implications of line tension as regards configurational stability are explored in Section 6.
2. ENERGY OF DELAMINATED FILMS

For many material systems of interest, the deflections of the film following delamination are observed to be of moderate size (see, e.g. Argon et al., 1989a, b), which suggests framing the analysis within the classical von Kármán theory of moderate deflections of a plate. We begin by investigating the role played by the various contributions to the energy of the film, as computed from von Kármán theory. We consider an infinite thin film of constant thickness \( h \) bonded to a substrate occupying the half-space \( x_3 \leq h/2 \) (Fig. 1). The film is in a state of residual biaxial compression and is debonded over a region \( \Omega \), where it buckles out of its plane to form a blister. The region \( \Omega \) has boundary \( \Gamma \) with outward unit normal \( \mathbf{n} \). We shall restrict our attention to bounded domains and presume both \( \Omega \) and \( \Gamma \) to be as regular as needed. Requiring \( \Omega \) to be open and bounded and \( \Gamma \) to be Lipschitz-continuous suffices for most mathematical purposes.

Let \( \mathbf{r} : \Omega \to \mathbb{R}^3 \) be the position vector of points on the midsurface of the film after deformation. The in-plane displacements and out-of-plane deflections of the film are defined as

\[
\begin{align*}
    u_\alpha &= r_\alpha - x_\alpha, \\
    w &= r_3,
\end{align*}
\]

respectively, where Greek indices range from 1 to 2. In von Kármán’s plate theory, the membrane and bending strains are defined as

\[
\begin{align*}
    \varepsilon_{\alpha\beta} &= \varepsilon_{(\alpha,\beta)} + w_{,\alpha}w_{,\beta}/2 - \varepsilon_{\alpha\beta}^\theta, \\
    \chi_{\alpha\beta} &= w_{,\alpha\beta},
\end{align*}
\]

where \( \varepsilon_{(\alpha,\beta)} = (u_{,\alpha} + u_{,\beta})/2 \) are the components of the symmetric gradient of the in-plane displacements, and \( \varepsilon_{\alpha\beta}^\theta \) are the eigenstrains in the film, i.e. the uniform strains the film would undergo if released from the substrate. The second term of (2) is nonlinear and couples, to leading order, the out-of-plane deflections to the in-plane deformations. The membrane and bending energy densities are then postulated to be isotropic quadratic functions of the membrane and bending strains, respectively, i.e.

\[
W^m = \frac{C}{2}[(1-v)\varepsilon_{\alpha\beta}\varepsilon_{\alpha\beta} + v(\varepsilon_{\gamma\gamma})^2],
\]

where

\[
W^b = 
\]

are the membrane and bending energy densities.

The problem of finding equilibrium fields, typically subject to Dirichlet and Neumann conditions on \( \Gamma \). This...

For fields rendering...

The equilibrium functional can be considered...
Thin-film blisters

\[ W^b = \frac{D}{2} [(1 - v) w_{,\alpha \beta} w_{,\alpha \beta} + v(w_{,\gamma \gamma})^2], \]  

(5)

where

\[ C = \frac{Eh}{(1 - v^2)}, \quad D = \frac{Eh^3}{12(1 - v^2)} \]  

(6)

are the membrane and bending stiffnesses of the film, which are defined in terms of the Young’s modulus \( E \) and Poisson’s ratio \( v \) of the material. The membrane stresses and bending moments follow as

\[ N_{\alpha \beta} = \frac{\partial W^m}{\partial \varepsilon_{\alpha \beta}} = C[(1 - v) \varepsilon_{\alpha \beta} + v \gamma_{\gamma} \delta_{\alpha \beta}], \]  

(7)

\[ M_{\alpha \beta} = \frac{\partial W^b}{\partial \chi_{\alpha \beta}} = D[(1 - v) w_{,\alpha \beta} + v w_{,\gamma \gamma} \delta_{\alpha \beta}]. \]  

(8)

The membrane and bending energies of the delaminated portion of the film are obtained by integration of (4) and (5), respectively, with the result

\[ \Phi^m[r] = \int_\Omega W^m \, dx_1 \, dx_2, \]  

(9)

\[ \Phi^b[r] = \int_\Omega W^b \, dx_1 \, dx_2, \]  

(10)

and the total energy of the film follows as

\[ \Phi[r] = \Phi^m[r] + \Phi^b[r]. \]  

(11)

The problem is, therefore, to minimize \( \Phi[r] \) over some suitable space \( X \) of displacement fields, typically a reflexive Banach space of functions satisfying the essential boundary conditions

\[ u_a = 0, \]  

(12)

\[ w = 0, \quad w_{,n} u_a \equiv w_{,n} = 0 \]  

(13)

on \( \Gamma \). This defines the variational problem

\[ (P) : \quad \inf \{ \Phi[r], r \in X \}. \]  

(14)

For fields of sufficient differentiability, the equilibrium equations are obtained by rendering (11) stationary. The result is

\[ N_{\alpha \beta, \alpha \beta} = 0, \]  

(15)

\[ M_{\alpha \beta, \alpha \beta} - (N_{\alpha \beta} w_{,n})_{,\beta} = 0. \]  

(16)

The equilibrium equations (15) and (16) constitute the Euler equations of the energy functional (11). Because of the lack of convexity of the energy functional under consideration, however, it will prove advantageous to resort to direct methods in the
calculus of variations. These methods endeavor to minimize the energy functional directly, sidestepping any consideration of Euler’s equations.

The energy functional (11) can be manipulated so as to bring it into closer correspondence with energy functionals of the type considered by James and Kinderlehrer (1990). Making use of (15), the membrane energy (9) can be expressed as

$$\Phi^m[r] = \int_\Omega \frac{C}{2} \left[ (1 - \nu)e_{\alpha\beta}^m e_{\alpha\beta}^m + \nu(e_{\gamma\gamma}^m)^2 \right] \, dx_1 \, dx_2 - \int_\Omega \frac{C}{2} \left[ (1 - \nu)e_{\alpha\beta}^s e_{\alpha\beta}^s + \nu(e_{\gamma\gamma}^s)^2 \right] \, dx_1 \, dx_2,$$

(17)

where we have denoted

$$\hat{\varepsilon}_{\alpha\beta}^m = \frac{1}{2} \varepsilon_{\alpha\beta}^m \delta_{\alpha\beta}^\ast, \quad \hat{\varepsilon}_{\alpha\beta}^s = u_{(\alpha\beta)},$$

(18)

and u and w are subject to (15). Interestingly, membrane solutions relaxing $\Phi^m[r]$ to zero result in energy equipartition in the sense that the identity

$$\int_\Omega \frac{C}{2} \left[ (1 - \nu)e_{\alpha\beta}^m e_{\alpha\beta}^m + \nu(e_{\gamma\gamma}^m)^2 \right] \, dx_1 \, dx_2 = \int_\Omega \frac{C}{2} \left[ (1 - \nu)e_{\alpha\beta}^s e_{\alpha\beta}^s + \nu(e_{\gamma\gamma}^s)^2 \right] \, dx_1 \, dx_2$$

(19)

is satisfied by the minimizer.

The analogy to magnetostatics is now apparent. The energy of a ferromagnet is commonly assumed to consist of three parts (James and Kinderlehrer, 1990):

$$\Phi[u, m] = \int_\Omega \nabla m \cdot A \cdot \nabla m \, dx_1 \, dx_2 + \int_\Omega \phi(m) \, dx_1 \, dx_2 + \int_\Omega \frac{1}{2} |\nabla u|^2 \, dx_1 \, dx_2$$

(20)

where $m$ is the magnetization, $u$ the magnetostatic potential, and A a symmetric tensor. The first term in (20) is the exchange energy, the second the anisotropy energy and the third the magnetostatic energy. A critical aspect of the anisotropy energy is that the function of $\phi(m)$ has multiple wells, corresponding to preferred directions for the magnetization. The magnetization and potential are subject to the constraint

$$\nabla \cdot (\nabla u + m) = 0,$$

(21)

which is a consequence of Maxwell’s equations. Inspection of (17) and (20) reveals a similar structure of the energy functional in both cases, with the slope $\nabla w$ playing the role of the magnetization $m$, and the in-plane displacements $u$ playing the role of the potential $u$. The exchange energy, which involves derivatives of $m$, has its counterpart in the bending energy, which likewise involves derivatives of $\nabla w$. The anisotropy energy has its analog in the first term of (17). Similarly to its magnetostatic counterpart, the corresponding energy density has multiple wells, which determine preferred values of the slope $\nabla w$, as demonstrated in subsequent sections. Finally the second term of (17) plays the role of the magnetostatic energy in (20), and constraint (15) is analogous to (21).

Connections can also be established with theories of liquid crystals. An incomplete minimization of the film’s energy can be effected simply by setting the in-plane displacements $u_\alpha = 0$ throughout $\Omega$, a choice which is trivially consistent with the boundary condition (12). This leads to the constrained functional

$$\Phi^m[w] = \int_\Omega \frac{C}{2} \left[ (1 - \nu)e_{\alpha\beta}^m e_{\alpha\beta}^m + \nu(e_{\gamma\gamma}^m)^2 \right] \, dx_1 \, dx_2,$$

where, for simplicity,$\hat{\varepsilon}_{\alpha\beta}^m = \varepsilon_{\alpha\beta}^m \delta_{\alpha\beta}^\ast$. A c

Let $a$ be a characteristic length, the limit of $h \to 0$ becomes

which defines a

Inspection of the second-order deformation tensor $m$ in first order only parameter and conventional expansion: the reduced vanishing in some approximative boundary layer.

An essential requirement is that the membrane energy be valued function; and only if $W^m$ would be not met may
\[
\Phi[w] = \int_\Omega \left[ \frac{C}{2} (1 - \nu^2)(\varepsilon^*)^2 + \frac{1}{4} (|\nabla w|^2 - 2(1 + \nu)\varepsilon^*) \right] \, dx_1 \, dx_2 \\
+ \int_\Omega \frac{D}{2} \left[ (1 - \nu)\varepsilon_{\alpha\beta} w_{,\alpha\beta} + \nu (w_{,\alpha})^2 \right] \, dx_1 \, dx_2, 
\]

(22)

where, for simplicity, we have assumed the eigenstrains to be isotropic, i.e. of the form \( \varepsilon_{\alpha\beta}^* = \varepsilon^* \delta_{\alpha\beta} \). A closely related functional has been considered by Avilés and Giga (1987) as a model for liquid crystals.

3. Thin-Film Limit

Let \( a \) be a characteristic dimension of \( \Omega \). The structure of the energy functional in the limit of \( h \to 0 \) may be revealed by normalizing all lengths by \( a \), whereupon (11) becomes

\[
\bar{\Phi}_\varepsilon[\tilde{r}] = \int_\Omega \left[ \bar{W}^{(m)} + \varepsilon^2 \bar{W}^{(h)} \right] \, d\tilde{x}_1 \, d\tilde{x}_2, 
\]

(23)

where a superimposed \( \sim \) denotes normalization by \( a \) and \( \varepsilon \equiv h/a \). Problem (P) thus becomes

\[
(P_\varepsilon): \quad \inf \{ \bar{\Phi}_\varepsilon[\tilde{r}], \tilde{r} \in \bar{X} \},
\]

(24)

which defines a one-parameter family of variational problems in the parameter \( \varepsilon \).

Inspection of the energy functional (23) reveals that the bending term contains second-order derivatives of \( w \), while the membrane term involves derivatives up to first order only. In addition, in the thin-film limit of interest here, \( \varepsilon \) becomes a small parameter and the bending energy singularity perturbs the membrane energy. The conventional approach to problems of this nature is to attempt a matched asymptotic expansion: the outer solution relaxes the membrane energy and, therefore, satisfies the reduced variational problem

\[
(P^{m}): \quad \inf \{ \Phi^{m}[r], r \in X^{(m)} \}
\]

(25)
in some appropriate configuration space \( X^{(m)} \). The inner solution is obtained by fitting boundary layers at points of slope discontinuity of the membrane deflections.

An essential difficulty which arises in carrying out this program is the fact that the membrane energy \( \Phi^{m} \) is a nonconvex functional of the deflections \( w \). Since \( w \) is a scalar-valued function of position and \( W^{(m)} \) depends on \( \nabla w \) and not on \( w \), \( \Phi^{m} \) is convex if and only if \( W^{(m)} \) is a convex function of \( \nabla w \) (Dacorogna, 1989). That this condition is not met may be exhibited simply by setting \( \nabla w = 0 \), whereupon \( W^{(m)} \) reduces to

\[
W^{(m)} = \frac{C}{2} \left[ (1 - \nu^2)(\varepsilon^*)^2 + \frac{1}{4} (|\nabla w|^2 - 2(1 + \nu)\varepsilon^*) \right],
\]

(26)
which is clearly nonconvex (Fig. 2). We remark in passing that $W^m$ is a convex function of the displacement gradients $\mathbf{V}u$, and, hence, $\Phi^m$ is a convex functional of $u$. In addition, $\Phi^m$ is coercive in $u$ over $H_0^1(\Omega; R^2)$, and, consequently, the in-plane displacements are uniquely defined once the deflections $w$ are specified (see, e.g., Marsden and Hughes, 1983; Dacorogna, 1989). Finally, we may note that, because $w$ is a scalar, one need not differentiate between convexity and quasiconvexity of $W^m$ as a function of $w$.

Nonconvex variational problems arise in the areas of structural optimization, phase transitions and homogenization, and the literature on the subject is presently quite extensive [see the review of Kohn and Strang (1986)]. The implications of the lack of convexity of $W^m$ are profound. For instance, the infimum of the membrane energy is generally not attained by any deflection $r$. With some ingenuity, however, it is sometimes possible to find minimizing sequences $r_j$ of deflections for which the energy $\Phi^m[r_j]$ attains values that are arbitrarily close to the infimum. Examples of minimizing sequences for the membrane energy are given in Section 4. Knowledge of these minimizing sequences, which invariably involve increasingly finer features, is desirable as they often reveal useful insights into the development of microstructures. Minimizing sequences can also be taken as a basis for predicting effective macroscopic behavior. This approach has been successfully applied in theories of ideal membranes (Fipkin, 1986a,b), martensitic transformations (Ball and James, 1987), ferromagnetism (James and Kinderlehrer, 1990) and magnetostriction (James and Kinderlehrer, 1993).

In effecting the minimization (25), one regards the film as an ideal membrane. Since no bending stiffness is attributed to ideal membranes, the minimizing sequences of (25) can be expected to be very simple.

Consideration of a variety of edge conditions permits more meaningful boundary conditions, if the folding problem is treated within a mathematical framework. Avilés and a slope discontinuity gives them a better perspective. Mathias, Avilés and a tension points to a possible bifurcation of solutions.

Some of these solutions can be effectively approximated by minimizing energy and the compliances of the membrane. An analysis leads to the conclusion of Section 5.7.

![Fig. 2. Membrane energy density $W^m$ as a function of $\mathbf{V}w$ ($\mathbf{V}u = 0, v = 0.2, \varepsilon^* = 0.012$).]
a convex functional of \( u \) in-plane (see, e.g., at, because of \( W^m \) vicinity, phase transition, quite energy of the lack energy even, it is the energy minimizing ge of these desirable areas. Microscopic membranes [87], ferro-James and boundary. Since sequences

of (25) can exhibit sharp edges across which the normal jumps discontinuously. Consideration of bending modifies the solution locally so as to round off the sharp edges over widths which decrease to zero as \( \varepsilon \to 0 \). Consideration of bending also permits modifying the solution within a boundary layer so as to satisfy the slope boundary condition \( w_r = 0 \). An additional effect of bending is to limit the fineness of the folding patterns which may be adopted by the film.

The treatment of these effects can be effectively simplified by regarding lines of slope discontinuity in the membrane solutions as sharp interfaces and according to them a bending energy per unit length, or line tension. There is a well-developed mathematical literature concerning sharp-interface limits of nonconvex variational problems regularized by higher-order gradients (see, e.g., Modica, 1987; Sternberg, 1988). Applications of such concepts to related problems have been pursued by Aviles and Giga (1987), and by Kohn and Muller (1992). The concept of line tension permits the assignment of a well-defined bending energy to membrane solutions. The problem is then to determine the membrane solution with the least possible bending energy. This defines a new variational problem, say \((P_0)\), which effectively selects among all possible solutions of \((P^m)\) one, \( r_0 \), with the least bending energy. The variational problem \((P_0)\) was termed by de Giorgi the \( \Gamma \)-limit of the complete problem \((P)\) as \( \varepsilon \to 0 \). Constructions for determining the preferred membrane solution \( r_0 \) are proposed in subsequent sections. A boundary layer analysis leading to the introduction of line tension at sharp edges is given in Section 5.2.

4. A TWO-VARIANT CONSTRUCTION

Some of the essential features of membrane energy minimizers are exhibited by the problem of a straight-sided semi-infinite film. Let the boundary coincide with the line \( x_2 = 0 \) and the film occupy the region \( x_2 \geq 0 \). Define the function \( \theta: R \to R \) as

\[
\theta(t) = \begin{cases} 
0, & \text{if } t \in [-l/2,0); \\
1, & \text{if } t \in [0,l/2), 
\end{cases}
\]

where \( l \) is some reference length parameter. Thus, \( \theta \) is a piece-wise constant periodic function of period \( l \) taking the value 0 over the interval \([-l/2,0)\) and 1 over the interval \([0,l/2)\) (Fig. 3). Let \( w \) be such that \( w(0,0) = 0 \) and

\[
\nabla w = [1 - \theta(x_1)] k \begin{pmatrix} 1/\sqrt{2} \\ 1/\sqrt{2} \end{pmatrix} + \theta(x_1) k \begin{pmatrix} -1/\sqrt{2} \\ 1/\sqrt{2} \end{pmatrix}.
\]

Thus the deflection \( w \) has a slope \( k \) alternating at \( 45^\circ \) and \( 135^\circ \) to the \( x_1 \)-axis over the strips \(-l/2 \leq x_1 < 0\) and \( 0 \leq x_1 < l/2\) labeled \( A \) and \( B \), respectively, in Fig. 3. The various facets in the deflection pattern meet at sharp edges of slope \( k/\sqrt{2} \). Evidently, the values of the deflection gradient \( \nabla w \) at \( x_1 = 0^\pm \) are

\[
\nabla w^\pm = k \begin{pmatrix} \mp 1/\sqrt{2} \\ 1/\sqrt{2} \end{pmatrix},
\]
and their contribution to the strain tensor is

\[ \frac{1}{2} (\mathbf{\nabla} \varepsilon \otimes \mathbf{\nabla} \varepsilon) = \frac{k^2}{4} \begin{pmatrix} 1 & \mp 1 \\ \mp 1 & 1 \end{pmatrix}. \]  

(30)

In addition, introduce an in-plane displacement field such that

\[ \mathbf{u} = [1 - \theta(x)] \frac{k^2}{2} \begin{pmatrix} 0 & 0 \\ -1 & 0 \end{pmatrix} + \theta(x) \frac{k^2}{2} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}. \]  

(31)

It is readily verified that these displacement gradients define a compatible displacement field. The corresponding strain tensor is piece-wise constant and takes the values

\[ (\nabla^s \mathbf{u})^\pm = \frac{k^2}{4} \begin{pmatrix} 0 & \pm 1 \\ \pm 1 & 0 \end{pmatrix}, \]  

(32)

where \( \nabla^s \mathbf{u} \) is the tensor of components \( u_{(a,\beta)} \).
Assume now that the eigenstrains are isotropic, i.e., $e_{ss}^* = \varepsilon^* \delta_{ss}$. Then, inspection of (28) and (31) reveals that the total strain tensor (2) can be made to vanish identically by setting

$$k = 2\sqrt{\varepsilon^*}.$$  \hspace{1cm} (33)

This choice renders the membrane energy density $W^m$ zero everywhere. Borrowing from the terminology commonly used in the crystallographic theory of martensite, a pair $(\nabla u, \nabla w)$ of displacement and deflection gradients will be said to define a variant if the corresponding membrane energy density is zero. Evidently, the displacement field defined in the foregoing contains two variants, labeled $A$ and $B$ in Fig. 3.

The two-variant construction just described minimizes the bulk energy of the film. The difficulty, of course, is that the displacement field so defined violates the boundary conditions (12). The deflection boundary condition can be satisfied by allowing the boundary to become wavy. Indeed, setting $w = 0$ in (28) gives the curve

$$x_2 = \begin{cases} -x_1, & \text{if } x_1 \in [-l/2, 0]; \\ x_1, & \text{if } x_1 \in [0, l/2], \end{cases} \hspace{1cm} (34)$$

which can be continued by periodicity to the entire real line. Let $\Gamma_1$ denote the boundary so defined. Evidently, by replacing $\Gamma$ by $\Gamma_1$ the deflections $w$ (28) satisfy the requisite boundary condition. By contrast, the in-plane displacements (31) remain in violation of the boundary condition $\partial u / \partial n = 0$ on $\Gamma_1$. Indeed, integration of (31) gives

$$u_1 = C_1, \quad u_2^+ = C_2 \pm \frac{k^2}{2} x_1, \hspace{1cm} (35)$$

where $C_\ast$ are integration constants. The choice $C_\ast = 0$ yields $u_\ast = 0$ at the origin.

For a similar problem concerning martensite twinning, Ball and James (1987) have shown that the boundary conditions can be accommodated by introducing a boundary layer, the width and energy of which go to zero as the fineness of the microstructure is allowed to become vanishingly small. Guided by this observation, we introduce folding patterns of increasing fineness by setting

$$\nabla w_j = [1 - \theta(jx_1)] k \begin{pmatrix} 1/\sqrt{2} \\ 1/\sqrt{2} \end{pmatrix} + \theta(jx_1) k \begin{pmatrix} -1/\sqrt{2} \\ 1/\sqrt{2} \end{pmatrix}, \hspace{1cm} (36)$$

$$\nabla u_j = [1 - \theta(jx_1)] \frac{k^2}{2} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} + \theta(jx_1) \frac{k^2}{2} \begin{pmatrix} 0 & 0 \\ -1 & 0 \end{pmatrix}, \hspace{1cm} (37)$$

where $j = 1, 2, \ldots$. In addition, define the boundary $\Gamma_j$ by the condition $w_j = 0$. Evidently, the boundaries $\Gamma_j$ undulate with an increasingly fine wavelength. Let $\Omega_j$ be the domain bounded by $\Gamma_j$. The complement $\Omega - \Omega_j$ consists of the triangular regions labeled $C$ in Fig. 3. The two-variant displacement field can be extended to all of $\Omega$ by setting $w = 0$ and

$$\nabla u = \frac{k^2}{2} \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \hspace{1cm} (38)$$
in $\Omega_1 \cup \Omega_2$. The displacement field so constructed is compatible over $\Gamma$. A simple calculation gives the energy of the film per unit length of the boundary $\Gamma$ as

$$
\frac{1}{L} \Phi^m[r_j] = \frac{I}{4j} C (1 - v)(\epsilon^*)^2.
$$

(39)

Evidently, the membrane energy of the film can be made arbitrarily small by letting $j \to \infty$, i.e., by increasing the fineness of the folding pattern. Consequently, the sequence of displacement fields $r_j$ constitutes a minimizing sequence.

It is interesting to note that, because of the lack of sequential weak lower semi-continuity of the membrane energy functional $\Phi^m[r]$, the limiting displacement field $r_j \to r$ is not an energy minimizer. Indeed, for any square $D \in \Omega$ of area $|D|$, it is easily verified that

$$
\frac{1}{|D|} \int_D \nabla u_j \, dx_1 \, dx_2 \to 0
$$

(40)

$$
\frac{1}{|D|} \int_D \nabla w_j \, dx_1 \, dx_2 \to \begin{pmatrix} 0 \\ \frac{2}{k} \end{pmatrix}
$$

(41)

which shows that

$$
\nabla u_j \to 0
$$

(42)

$$
\nabla w_j \to \begin{pmatrix} 0 \\ \frac{2}{k} \end{pmatrix}
$$

(43)

But the energy density of the limiting displacements is

$$
\frac{1}{|D|} \Phi^m = \frac{C}{2} (\epsilon^*)^2
$$

(44)

and, therefore, the limiting displacement field is not a minimizer.

Several conclusions can be drawn from the above construction. Firstly, it is observed that, by suitably folding the film, its bulk membrane energy can be reduced to values arbitrarily close to zero. In fact, a combination of two variants suffices to relax the bulk membrane energy of the film completely. Additionally, it is found that satisfaction of displacement boundary conditions generally requires the introduction of a boundary layer. However, the thickness and membrane energy of the layer can be made arbitrarily small by refining the folding pattern. Finally, wavy boundaries are energetically favorable in as much as they help to accommodate the internal folding of the membrane, thus reducing the energy of the boundary layer.

5. GENERAL DOMAINS

It would seem natural to attempt a generalization to arbitrary domains of the folding construction given in the preceding section. We have found, however, that good agreement with experiment in the case of the unstressed film is obtained only when the conditions for the energy functional $\Phi$ are specified properly.

5.1. Membrane energy

Consider now the analogy of the eigenstrain of the membrane functional $\Phi$

$$
\Phi[w] = C \frac{e^*}{2}
$$

where

$$
\int_D \nabla w \cdot \nabla u \, dx_1 \, dx_2
$$

It is apparent that

$$
\int_D \nabla w \cdot \nabla w \, dx_1 \, dx_2
$$

(45), namely

$$
\int_D \frac{1}{2} \nabla w \cdot \nabla w \, dx_1 \, dx_2
$$

(46)

Clearly, the results of the previous section apply in the present case.

Evidently, a generalization of the above result is possible.

Equation (46)

is obtained as the energy expression between two wavefronts, $u_b$ and $u_{b+}$, of a strained plastic bar.

A difficult case is that of a problem with a plate simply supported on the boundaries $y = \pm b$ constituting...
good agreement with observation is obtained by recourse to a simplified theory, which we proceed to develop in this section. The theory consists of setting the in-plane displacements \( u \) to zero and effecting a—necessarily partial—relaxation of the resulting energy functional of \( w \), namely (22). This simplification leads to a simple construction for computing the deflections of blisters of arbitrary shape. In addition, the simplified theory affords some quantitative predictions of the shape of blisters, as discussed in Section 6.

5.1. Membrane solutions

Consider now an open and bounded domain \( \Omega \) having a smooth boundary \( \Gamma \). Let the eigenstrains be isotropic, i.e. of the form \( \varepsilon_{\alpha\beta} = \varepsilon^* \delta_{\alpha\beta} \). By setting \( u = 0 \), the energy functional \( \Phi \) reduces to (22), which can be rewritten as

\[
\Phi[w] = C_0 + \int_{\Omega} \frac{C}{8} (|\nabla w|^2 - k^2)^2 \, dx_1 \, dx_2 + \int_{\Omega} \frac{D}{2} [(1 - \nu)w_{,\alpha\beta}w_{,\alpha\beta} + \nu(w_{,\gamma\gamma})^2] \, dx_1 \, dx_2,
\]

(45)

where

\[
C_0 = \int_{\Omega} \frac{C}{2} (1 - \nu^2)(\varepsilon^*)^2 \, dx_1 \, dx_2
\]

(46)

\[
k = \sqrt{2(1 + \nu)\varepsilon^*}.
\]

(47)

It is apparent from (45) that the energy \( \Phi \) cannot be reduced beyond the value \( C_0 \).

We start by looking for minimizers of the membrane part of the energy functional (45), namely

\[
\Phi^m[w] = C_0 + \int_{\Omega} \frac{C}{8} (|\nabla w|^2 - k^2)^2 \, dx_1 \, dx_2.
\]

(48)

Evidently, \( \Phi^m \) is minimized by deflections \( w \) such that

\[
|\nabla w| = k, \quad \text{a.e. in } \Omega,
\]

(49)

\[
w = 0 \quad \text{on } \Gamma.
\]

(50)

Equation (49) is the Eikonal equation of geometrical optics. This affords an analogy between membrane solutions and optics in which level contours of \( w \) play the role of wavefronts, and the lines of steepest descent are the counterpart of rays. Equations (49) and (50) are also identical to those satisfied by the stress potential of an ideally plastic bar subjected to torsion.

A difficulty that arises in dealing with boundary value problem (49) and (50) is that it does not determine a unique solution. Consider, for instance, the one-dimensional problem and let \( \Omega \) coincide with the interval \([-b, b]\). Then, any polygonal line supported on this interval, consisting of segments of slope \( w' = \pm k \), and vanishing at \pm b constitutes a solution of (49) and (50) [Fig. 4(a)]. Likewise, in two dimensions,
the upper envelope of any collection of cones of slope $k$ supported in $\Omega$ is a solution of (49) and (50) [Fig. 4(b)]. However, among all possible solutions of (49) and (50), we are primarily interested in those which are as close as possible to minimizers of the complete potential (45) as $\epsilon = h/a \to 0$. This rules out deflection patterns which, while being solutions of (49) and (50), give rise to high energies when corrected for bending. Thus, in effect, we shall let bending select a preferred membrane solution. To this end, we must first have the means of computing the bending energy of possibly nonsmooth membrane deflections. This may be accomplished by a boundary layer analysis of sharp edges, as demonstrated next.

5.2. Local analysis of sharp edges

A particularly elegant and concise derivation of the energy of sharp interfaces has been given by Modica (1987) and is subsequently adapted to the present setting. Let the axes $x_1$ and $x_2$ be aligned with the transverse and parallel directions to the edge at the point under consideration (Fig. 5). Let $k_2$ be the slope of the film along the edge. Within the boundary layer one has, to leading order, $w_2 \approx k_2$ and $w_{11}$ dominates over $w_{12}$ and $w_{22}$.

where $k_1 = k_2 = 1/N$.

It should be recalled that $u_1$ is the indentation therefore, $u_1 = 0$.

A first integer

where we

Multiplyin

Integrating over $w_{12}$ and $w_{22}$.

or $W^m = 1$.

By virtue of

At first glance identifying
over $w_{12}$ and $w_{22}$. Consequently, the energy per unit length of the layer can be approximated as

$$\frac{\Phi}{L} = T = \int_{-\infty}^{\infty} \left[ \frac{C}{2} \left( \frac{1}{4} (w''^2 - k_1^2) \right)^2 + \frac{h^2}{12} w''^2 \right] dx_1,$$

(51)

where $k_1 = \sqrt{k^2 - k_2^2}$ and primes are used to denote differentiation with respect to $x_1$. It should be noted that $T$ represents an energy per unit length of the edge and, therefore, may be regarded as a line tension. The Euler equation associated with (51) is

$$-C \left[ \frac{1}{2} (w''^2 - k_1^2) w' \right] + D w''' = 0.$$

(52)

A first integral of this ordinary differential equation is

$$-C \left[ \frac{1}{2} (w''^2 - k_1^2) w' \right] + D w''' = 0,$$

(53)

where we have used the condition that $w' \to k_1$ and $w'' \to 0$ away from the edge. Multiplying (53) by $w''$ gives the identity

$$\left\{ -\frac{C}{2} \left[ \frac{1}{4} (w''^2 - k_1^2) \right] + \frac{D}{2} w''^2 \right\}' = 0.$$

(54)

Integrating with respect to $x_1$ and imposing that $w' \to k_1$ and $w'' \to 0$ away from the edge finally gives

$$\frac{C}{2} \left[ \frac{1}{4} (w''^2 - k_1^2) \right] = \frac{D}{2} w''^2,$$

(55)

or $W^m = W^b$. This proves the equipartition of the energy within the boundary layer. By virtue of this equipartition, the line tension (51) can be written as

$$\frac{\Phi}{L} = T = 2 \int_{-\infty}^{\infty} \frac{C}{2} \left[ \frac{1}{4} (w''^2 - k_1^2) \right] dx_1.$$

(56)

At first glance, it seems that the deflection $w(x_1)$ across the boundary layer is required in order to compute $T$. The computation of $w$, however, is rendered unnecessary by identifying $w'$ as the independent variable. Indeed, setting $\xi = w'$ gives $dx_1 = d\xi/w''$. 

\[ \text{Thin-film blisters} \]
Eliminating $w$ with the aid of (55), the right-hand side of (56) reduces to the elementary integral

$$T = \frac{1}{2} \sqrt{CD} \int_{-k_1}^{k_1} |\xi^2 - k_1^2| \, d\xi = \frac{3}{2} \sqrt{CD} k_1^3,$$  \quad (57)

which completes the calculation of $T$. At points on the boundary $\Gamma$ of the domain of the blister, the boundary layer is one-sided [Fig. 5(b)], and coincides with one half of the boundary layer at an interior edge [Fig. 5(a)]. In addition, $k_1 = k$ on the boundary. As a consequence, the energy per unit length, or line tension, of the boundary takes the constant value

$$T = \frac{1}{2} \sqrt{CD} k^3.$$  \quad (58)

Finally, a conventional width $l$ can be assigned to the boundary layer by estimating the curvature within the layer as $2k_1/l$ and requiring that

$$T = D \left( \frac{2k_1}{l} \right)^2 l.$$  \quad (59)

Inserting (57) into this definition gives

$$l = \sqrt{\frac{3}{D}} h k_1.$$  \quad (60)

As expected, the edge width scales with $h$ and, consequently, becomes vanishingly small as $h \to 0$. It is also noted from the relation $k_1 = \sqrt{k^2 - k_3^2}$ that the width of the edge is a function of its slope $k_2$. In particular, the width attains its minimum value when $k_2 = 0$ and diverges to infinity when $k_2 \to k$.

### 5.3. Envelope construction

The bending energy of membrane solutions can now be estimated as follows. In the thin film limit $\varepsilon = h/a \to 0$, the boundary layer analysis of the preceding section applies and the bending energy of the film is given by

$$\Phi_0 = \frac{1}{2} \sqrt{CD} k^3 L + \int_L \frac{3}{2} \sqrt{CD} k_1^3 \, ds,$$  \quad (61)

where $L$ is the length of the boundary $\Gamma$, $L$ denotes the collection of interior edges in the membrane solution, and $s$ is the arc-length measured along $L$. Conveniently, (61) can be expressed as an area integral over $\Omega$ of the form

$$\Phi_0 = \int_\Omega \frac{1}{2} \sqrt{CD} |w_{,\alpha\beta} w_{,\alpha\beta}| \, dx_1 \, dx_2.$$  \quad (62)

Indeed, at regular points of the membrane solution differentiation of (49) gives $w_{,\alpha\beta} w_{,\beta} = 0$, and, therefore, the only contribution to (62) comes from the edges. At an interior edge, however, the dominant component of $\nabla w$ is $w_{,11}$, which takes the value $\pm 2k_1 \delta(x_1)$. The conditions, as argued by integrating the term in (61).

As argued to select that

The preference

subjected to

Mathematical

of problem

of Giorgi and

Giorgi's the

and Stern

In two-di

that with the

this is simple

In two-di

energy $\Phi_0$ is

Equivalent to

Nadai's sar

for an ideal

as the envelope

the preference

5.4. Bending

Once the

5.2. An alt

the equilibri

responding
\[ \pm 2k_{1} \delta(x_1) \] Here \( \delta(x_1) \) denotes the Dirac delta, and we have adopted the local axes employed in the boundary layer analysis of the preceding section. Under these conditions, one computes \(|w_{\alpha}w_{\beta}w_{\gamma}| = 2k_{1}^{2}\delta(x_1)\), and the second term in (61) follows by integration over \( \Omega \). The boundary \( \Gamma \) can be treated similarly, leading to the first term in (61).

As argued in the foregoing, of all possible solutions of (48) one may expect bending to select that containing the least residual energy \( \Phi_0 \), which for \( \veps \to 0 \), is given by (62). The preferred membrane solution satisfies the problem

\[ (P_0): \quad \inf \{ \Phi_0(r), \, r \in X^m \} \]

subjected to

\[ |\nabla w| = k \quad \text{a.e. in } \Omega, \quad w = 0 \quad \text{in } \Gamma. \]

Mathematically, the variational problem \( (P_0) \) just defined may be regarded as a limit of problem \( (P_2) \) as the thin-film limit \( \veps \to 0 \) is approached. DE GIORGI (1975) and DE GIORGI and FRANZONI (1975), in pioneering work, developed a mathematically rigorous method for effecting this limit, which he called the \( \Gamma \)-limit. Applications of De Giorgi’s theory to phase transition problems have been pursued by MODICA (1987) and STERNBERG (1988).

In one-dimension, \( \Phi_0 \) simply counts the number of slope discontinuities in a deflection pattern such as shown in Fig. 4(a). The preferred membrane solution is, therefore, that with the least number of discontinuities, i.e. one consisting of two straight segments of slopes \( \pm k \) meeting at the center of the interval [Fig. 5(a)]. Interestingly, this is simply the upper envelope of all solutions of the Eikonal equation (49).

In two-dimensions, we conjecture that the membrane solution with the least bending energy \( \Phi_0 \) is, likewise, the upper envelope of all solutions of the Eikonal equation. Equivalently, the preferred membrane solution so defined can be computed as the upper envelope of all cones of slope \( k \) supported in \( \Omega \) [Fig. 4(b)]. This is, of course, Nadai’s sand-heap construction (NADAI, 1950) for determining the stress potential for an ideally plastic bar subjected to torsion. Yet another equivalent construction is as the envelope of all planes of slope \( k \) tangent to the boundary \( \Gamma \), which shows that the preferred membrane deflection defines a tangent-developable surface (STRUik, 1950). As discussed in Section 5.5, the folding patterns predicted by these constructions are in remarkably good agreement with observation, which lends empirical support to the present theory.

5.4. Bending and smoothing

Once the membrane solution has been computed, the effect of bending may be taken into account by fitting boundary layers to sharp edges, as discussed in Section 5.2. An alternative, albeit cruder, means for taking bending into account is to linearize the equilibrium equation about the membrane solution. The Euler equation corresponding to the energy functional (45) is

\[ -\nabla \cdot [(|\nabla w|^2 - k^2)\nabla w] + \frac{h^2}{12} \nabla^4 w = 0. \]
Let \( w_0 \) be the solution of problem (48). Then, a linearization of (65) about \( w_0 \) gives

\[
-2\nabla \cdot \left[ (\nabla w_0 \otimes \nabla w_0) \cdot \nabla (w - w_0) \right] + \frac{h^2}{12} \nabla^4 w = 0,
\]

(66)

where use has been made of the identity \( |\nabla w_0| = k \), \( \otimes \) denotes dyadic product, and we have neglected terms of \( O[|\nabla w - \nabla w_0|/k^2] \) or higher. Equation (66) can be further simplified as follows. Let \( p = \nabla w_0/k \) be the unit vector in the direction of \( \nabla w_0 \), and \( q \) the orthogonal unit vector which, together with the unit normal to the plane of the film, defines a right-handed orthonormal triad. With the notation, \( (\nabla w_0 \otimes \nabla w_0) \cdot \nabla (w - w_0) = k^2 (p \otimes p) \cdot \nabla (w - w_0) \), but the difference \( \nabla (w - w_0) \) only becomes appreciable within boundary layers, wherein \( \nabla (w - w_0) \cdot q \approx 0 \). Hence, we can write

\[
(\nabla w_0 \otimes \nabla w_0) \cdot \nabla (w - w_0) \approx k^2 (p \otimes p + q \otimes q) \cdot \nabla (w - w_0) = k^2 \nabla (w - w_0),
\]

since \( (p \otimes p + q \otimes q) \) coincides with the identity tensor. Inserting this identity into (66) gives

\[
-2\nabla^2 (w - w_0) + \frac{h^2}{12k^2} \nabla^4 w = 0.
\]

(67)

In principle, this equation is subject to the boundary conditions (13). Imagine, however, that the domain \( \Omega \) is bounded, and let \( r \) denote the radial distance to the origin. It is then expedient to extend the domain of (67) to the complete plane by setting \( w_0 = 0 \) outside \( \Omega \) and by replacing (13) by the conditions \( w \to 0 \) and \( w_r \to 0 \) at infinity. Then one Laplacian in (67) can be eliminated, with the result

\[
(1 - \lambda^{-2} \nabla^2) w = w_0,
\]

(68)

where we have defined

\[
\lambda = \sqrt{\frac{24}{h}}.
\]

(69)

Equation (68) shows that, under the assumptions of the analysis, the bending solution is obtained from the membrane solution by Laplacian smoothing. The solution of (68) can be written as

\[
w_c = \phi_c \ast w_0.
\]

(70)

Here, \( \ast \) denotes convolution and the mollifier \( \phi_c \) is given by

\[
\phi_c = \lambda^2 K_0(\lambda r),
\]

(71)

where \( K_0 \) is the Bessel function of the second kind and order zero. As expected, the mollifiers \( \phi_c \) define a delta sequence, i.e. \( \phi_c \to \delta \) in the thin limit \( \varepsilon \to 0 \) in the sense of distributions. Suggestively, the membrane solution \( w_0 \) is recovered as the thin film limit of \( w_c \), i.e. \( w_c \to w_0 \) exponentially away from the substrate.

(70) yields smoothed solutions of delaminations are negligibly small.

The smoothing of delaminations is predicted by (70) remains to be experimentally verified. The levels predicted are not excessive.

5.2. Consideration of experiment

For \( x_1 > 0 \),

The energy

Using definition

Comparision overestimate

5.5. Comparison of experiment

Blisters caused by the chemical stress of experimentally. Considerably the case of

\( E \approx 187 \) GPa characterizes.

In order
limit of \( w \), i.e. in the limit of \( \varepsilon \to 0 \). It should be noted, however, that, for finite \( \varepsilon \), (70) is not in strict compliance with boundary conditions (13). Instead, \( \phi \to 0 \) exponentially away from the boundary over distances of order \( \lambda^{-1} = h/k\sqrt{24} \). Thus, while (70) yields some partial delamination outside \( \Omega \), the layer over which this spurious delamination takes place shrinks to the boundary as \( \varepsilon \to 0 \), and may therefore be neglected to a first approximation.

The smoothing procedure just described provides a particularly convenient means of post-processing the membrane solution so as to account for bending effects. In deriving (70), however, various approximations have been introduced whose accuracy remains to be determined. As an accuracy assessment, we may compare the energy levels predicted by the smoothing procedure to the exact results derived in Section 5.2. Consider an interior sharp edge such as shown in Fig. 5(a). Then (68) reduces to

\[
\dot{w} - \lambda^{-2} w'' = w_0. \tag{72}
\]

For \( x_1 > 0 \), the solution to this equation which has a vanishing slope at the origin is

\[
w = A - k(x_1 + \lambda^{-1} e^{-\lambda x_1}). \tag{73}
\]

The energy per unit length of the edge is then computed as

\[
T = \frac{\sqrt{2}}{2} \sqrt{C} D k^3. \tag{74}
\]

Using definition (59), the width of the boundary layer is computed to be

\[
l = \sqrt{\frac{8}{3} \frac{h}{k}}. \tag{75}
\]

Comparison with the exact values (57) and (60) reveals that the linearized theory overestimates both the energy and the width of the boundary layer by roughly 6%.

5.5. Comparison with experiment

Blister types of the type considered in this paper have been extensively documented in the experimental literature. Figures 6(a) and 7(a) show two particularly well-developed blisters reported by ARGON et al. (1989a, b). In these tests, amorphous hydrogenated thin films of SiC were deposited on Si single crystal wafers using a plasma-assisted chemical vapor deposition (PACVD) process. The dependence of the residual compressive stress left in the coating on the ion bombardment energy is known experimentally. In the tests of ARGON et al. (1989a, b), the residual stress attains the considerable value of 2 GPa. The thickness of the film was measured to be 1.1 \( \mu \)m in the case of Fig. 7(a), and is presumably comparable in the case of Fig. 6(a). Taking \( E \approx 187 \) GPa and \( \nu \approx 0.2 \), the resulting compressive eigenstrain \( \varepsilon^* = 0.011 \) and the characteristic slope \( k = 0.15 \).

In order to test the ability of the theory to reproduce observed folding patterns
Fig. 6. (a) SiC/Si blister reported by Argon et al. (1989a). (b) Membrane solution by upper envelope construction. (c) Solution after bending correction.
when the domain of the blister is known, we have digitized the boundaries of the blisters reported by ARGON et al. (1989a, b) and applied to them the method of analysis developed in the preceding sections. The result of the upper envelope construction described in Section 5.3, giving the preferred membrane solution, is shown in Figs 6(b) and 7(b). It bears emphasis that, while the solution is evaluated on a square grid for purposes of graphical display, the method of solution is essentially analytical. As may be observed from these figures, the membrane solution exhibits sharp edges in the interior of the domain. Figures 6(c) and 7(c) show the same solution after correcting for bending by the method of smoothing developed in Section 5.4. Predictably, the sharp edges take on a rounded appearance upon smoothing. It is entertaining to compare the analytical solutions with the observed blisters fold by fold. The ability of the theory to reproduce, by a simple construction, intricate details of the observed folding patterns is quite remarkable.
Fig. 7. (a) SiC/Si blister reported by ARGON et al. (1989b). (b) Membrane solution by upper envelope construction. (c) Solution after bending correction.
A comparison of predicted and measured wavelengths of the boundary undulations affords a further test of the theory. We argue that, in the blisters reported by Argon et al. (1989a, b), and shown in Figs 6(a) and 7(a), the boundary undulations simply accommodate the interior folds, i.e. the boundary is, to a first approximation, a level contour of the folding pattern. Recent observations by Thouless (private communication) support this view. He tested a thin film of mica on an aluminum substrate into which he introduced a large and roughly circular blister by driving a screw through the substrate. When the screw was removed, the film folded into patterns reminiscent of those observed by Argon et al. (1989a, b), and the boundary receded so as to match the interior folding.

Under these circumstances, the wavelength of the boundary should be of the order of twice the width $l$ of an interior edge. For the SiC/Si system tested by Argon et al. (1989a, b), one has $h = 1.1$ $\mu m$ and $k = 0.15$, which, when inserted into (60), give
$l = 12.7 \mu m$. Consequently, the wavelength of the boundary is predicted to be of the order of $25.4 \mu m$, which, by simple inspection, appears consistent with observations [see Figs 6(a) and 7(a)].

6. Configurational Stability

The boundary layer analysis given in Section 5.2 permits one to accord a well-defined energy per unit length, or line tension, to the boundary of the blister. Next, we explore the consequences of line tension as regards the shape of the blister. To this end, we begin by considering an infinitesimal element of the boundary of length $ds$ and radius of curvature $\rho$ which advances by a distance $da$ normal to itself (Fig. 8). Before delamination, the energy density of the film is

$$W^* = C(1 + \nu)(\varepsilon^*)^2.$$  \hspace{1cm} (76)

The energy density of the film in the wake of the delamination front, outside the boundary layer, is

$$W_0 = \frac{C}{2} (1 - \nu^2)(\varepsilon^*)^2.$$  \hspace{1cm} (77)

In addition, the line tension (58) can be expressed directly in terms of the mismatch strain $\varepsilon^*$ by recourse to (6) and (47), with the result

$$T = \frac{1}{6\sqrt{3}} Ch[2(1 + \nu)\varepsilon^*]^{3/2}.$$  \hspace{1cm} (78)

Balance of energy requires that the energy released by the film during the delamination process be expended in increasing the energy of the boundary and in creating a new surface. The energy required for the latter purpose is determined by the surface energy $\gamma_c$ of the film/substrate interface. As is well-known from interfacial fracture mechanics, $\gamma_c$ generally depends strongly on the mode mixity at the delamination front, as measured by the phase angle $\psi$ [see the review of Hutchinson and Suo (1991)]. For a thin film on an elastic substrate, Hutchinson and Suo (1991) have determined the phase angle to be

$$\psi = \frac{\rho}{\rho_c}.$$  \hspace{1cm} (79)

This relation is obtained under the assumption that $\rho \to \infty$ in (79), and the straight boundary $\Gamma$ is present. In contrast, for a thin film on a brittle substrate to delaminate

$$\psi = \frac{\rho}{\rho_c},$$  \hspace{1cm} (80)

is attained when $\rho$ is finite. In this case, $\rho_c$ is found to be a function of the crack driving force, which is supercritical when $\rho_c$ is

Fig. 8. Element of a decohering boundary.
\[ \tan \psi = \frac{\sqrt{12M \cos \omega + h\Delta N \sin \omega}}{-\sqrt{12M \sin \omega + h\Delta N \cos \omega}}. \] (79)

Here, \( M \) is the bending moment normal to the boundary acting on the interior edge of the blister, \( \Delta N \) is the jump in the axial force normal to the boundary upon buckling of the thin film, and \( \omega \) is a function of the elastic constants of film and substrate. Since \( \nabla w = 0 \) along the edge of the blister, we have, in the present theory, \( \Delta N = 0 \). Under these conditions, (79) reduces to

\[ \tan \psi = -\cot \omega, \] (80)

and \( \psi \) remains constant along the boundary of the blister. Thus, variations in phase angle do not play a significant role in the present theory. The state of mode mixity characterized by (80) coincides with that which exists at the edge of a one-dimensional blister in the limit of vanishing buckling deflections, as well as in the case of a blister driven by moderate values of internal pressure (Hutchinson and Suo, 1991).

Balance of energy during delamination requires

\[ (W^* - W_0) \, ds \, da = \mathcal{G}_c \, ds \, da + T \frac{ds}{\rho} \, da, \] (81)

or, eliminating the differentials,

\[ W^* - W_0 = \mathcal{G}_c + \frac{T}{\rho}. \] (82)

Finally, inserting (76)–(78) into (82) yields

\[ \frac{C}{2} \frac{(1 + \nu)^2 (e^*)^2}{2} = \mathcal{G}_c + \frac{C}{6\sqrt{3}} \frac{[2(1 + \nu)(e^*)^{3/2}] h}{\rho}. \] (83)

This relation determines an equilibrium radius of curvature of the boundary. Taking \( \rho \to \infty \) in (83) the critical eigenstrain

\[ e^* = \sqrt{\frac{2\mathcal{G}_c}{C(1 + \nu)^2}} \equiv e_c^* \] (84)

is obtained. The significance of this result is that, for subcritical eigenstrains, \( e^* < e_c^* \), straight boundaries, such as found in straight-sided blisters, are stable. By way of contrast, for supercritical eigenstrains, \( e^* > e_c^* \), straight boundaries lose stability and delaminate spontaneously. Delamination stops when the critical radius of curvature

\[ \rho = \frac{2}{3} \sqrt{\frac{2}{3(1 + \nu)}} (e^*)^{3/2} \frac{(e^*)^{3/2}}{(e^*)^{2}} h \equiv \rho_c \] (85)

is attained. As expected, \( \rho_c \) diverges as \( e^* \to e_c^* \) from above. Interestingly, for \( e^* \gg e_c^* \), \( \rho_c \) is found to decrease as \( 1/\sqrt{e^*} \). For fixed \( e^* \), segments of the boundary having a subcritical radius of curvature, \( \rho < \rho_c \), are stable. Conversely, intervals having a supercritical radius of curvature \( \rho > \rho_c \) are unstable and will delaminate spon-
taneously until the critical curvature is attained. Thus, $\rho_\alpha$ sets an upper bound for the radii of curvature which can be adopted by the boundary of a blister.

In large blisters, the attainment of the critical radius of curvature may require the boundary to break into adjacent arcs of circles separated by cusps. A case in point is the so-called telephone-cord morphology (Fig. 9). One way in which this morphology can be induced is the following. Imagine a straight-sided blister of width $b$ subjected to increasing eigenstrains $\varepsilon^*$ as a result, say, of a drop in temperature. Assume that, initially, $\varepsilon^*$ is subcritical so that the straight configuration is stable. At some point during the cooling process, the blister loses stability and delaminates transversely into adjacent arcs of circle of the critical curvature, which increases steadily with $\varepsilon^*$ as the system is cooled further. In principle, it would appear that the resulting waviness could be arbitrarily out of phase on both sides of the blister. However, in order to develop a smooth midridge (Fig. 9), the arcs of circle on both sides of the blister must be staggered precisely as in the telephone-cord morphology.

A system tested by Thouless (private communication) exhibits the precise transition just described. The system consists of a thin film of mica on an aluminum substrate into which an essentially straight-sided blister is introduced by driving screws through the substrate [Fig. 10(a)]. The system is subsequently cooled, thus raising the value of the mismatch strain $\varepsilon^*$. At a critical temperature drop, the blister is observed to delaminate sideways into a telephone-cord morphology [Fig. 10(b)]. Although the delamination occurs in spurts, the general trend is towards arcs of increasing curvature with increasing $\varepsilon^*$, in accordance with (85).

The present theory thus suggests that the telephone-cord morphology arises in the supercritical regime as a consequence of the curvature constraint. It should be noted, however, that, in many instances, telephone-cord blisters grow at the tip. Then, the lateral delamination which is so clearly exhibited by Thouless’ experiment cannot easily be separated from the forward motion of the tip, and the blister appears to propagate forward in a meandering fashion.

7. SUMMARY AND DISCUSSION

We have developed a method of analysis of thin film blisters subjected to biaxial compression based on energy methods. Exploiting the perturbative character of bending
Fig. 10. Test of mica-Al system demonstrating the loss of stability of a straight boundary at a critical eigenstrain (Thouless, private communication). Top: (a) initial straight-sided blister. Bottom: (b) telephone-cord morphology induced by cooling.
in the thin-film limit, the analysis is split along the conventional lines of matched asymptotic expansions: the outer solution follows membrane energy minimization, which favors fine folding; the inner solution is dominated by bending. The effect of bending is manifold. It endows sharp edges in the membrane solution with a well-defined width and line tension. It determines the wavelength of the folding and boundary undulations. Among all possible membrane solutions, it selects one which contains the least edge energy. We have conjectured that this preferred membrane solution follows as the result of a simple envelope construction. The analytical solutions thus constructed are found to be in remarkable agreement with observations. Finally, consideration of line tension enables, in certain cases, quantitative predictions of the radius of curvature of the boundary, and provides a rationale for explaining the occurrence of telephone-cord morphologies.

In closing, some remarks on the range of applicability of the theory are in order. The boundary layer analysis given in Section 5.2 shows that folds have a characteristic width $l$. Bending thus introduces a size effect: blisters of a size smaller than $2l$ cannot contain folds and remain smooth; by contrast, blisters of a size much larger than $2l$ can fit fully developed boundary layers in their interior and, consequently, will exhibit a perturbation to fold. For small blisters, the treatment of bending as a singular perturbation is not justified. Bending dominates the response of the blister, which buckles much as beams or plates do. It seems, therefore, appropriate to refer to this range of behavior as the buckling regime. By contrast, the essential features of the folding patterns adopted by large blisters are governed by the membrane energy of the film. Consequently, this range of behavior may be thought of as a folding regime. It is precisely in this latter regime, characterized by large blisters and fine folding, that the theory developed here applies.

ACKNOWLEDGEMENTS

This work has been funded by the NSF through Brown University's Materials Research Group on "Micro-Mechanics of Failure Resistant Materials". We are grateful to M. D. Thouless for useful discussions, and for making available to us the photographs shown in Fig. 10. We also wish to thank J. W. Hutchinson for some useful comments and suggestions.

REFERENCES


of matched imization, he effect of with a well-olding and one which membrane mytical solutions. predictions explaining

e in order. characteristic 2I cannot greater than 2I will exhibit angular perich buckles its range of the folding of the film regime. It is ng, that the


