# Folding Patterns in Partially Delaminated Thin Films

David Bourne, Sergio Conti and Stefan Müller

**Abstract** Michael Ortiz and Gustavo Gioia showed in the 90s that the complex patterns arising in compressed elastic films can be analyzed within the context of the calculus of variations. Their initial work focused on films partially debonded from the substrate, subject to isotropic compression arising from the difference in thermal expansion coefficients between film and substrate. In the following two decades different geometries have been studied, as for example anisotropic compression. We review recent mathematical progress in this area, focusing on the rich phase diagram of partially debonded films with a lateral boundary condition.

# 1 Introduction

Elastic films deposited on a substrate are often subject, after thermal expansion, to compressive strains which are released by debonding and buckling, generating a variety of microstructures. The work of Michael Ortiz and Gustavo Gioia in the 90s [1, 2] opened the way for the use of the tools of calculus of variations in the study of these structures. Their starting point was the Föppl-von Kármán plate theory, as given in (4) below. One of their insights was that the key nonconvexity which gives rise to the microstructure can be understood in terms of the out-of-plane displacement alone, leading after some rescalings to the Eikonal functional, as given in (1) below. This functional contains a term of the form  $(|Dw|^2 - 1)^2$ , where w is the normal

D. Bourne

S. Conti (⊠) · S. Müller Institut Für Angewandte Mathematik, Universität Bonn, Bonn, Germany e-mail: sergio.conti@uni-bonn.de

S. Müller

© Springer International Publishing Switzerland 2016

K. Weinberg and A. Pandolfi (eds.), *Innovative Numerical Approaches* for Multi-Field and Multi-Scale Problems, Lecture Notes in Applied and Computational Mechanics 81, DOI 10.1007/978-3-319-39022-2\_2

Department of Mathematical Sciences, Durham University, Durham, UK e-mail: david.bourne@durham.ac.uk

e-mail: stefan.mueller@hcm.uni-bonn.de

displacement, which favours deformations with the property that the gradient of w is approximately a unit vector, independently of the orientation. Since the film is still bound to the substrate at the boundary of the debonded region, the appropriate boundary condition is w = 0, which prescribes that the average over  $\Omega$  of the gradient of w vanishes. Therefore the resulting low-energy deformations have gradient Dw oscillating between different values. As in many nonconvex variational problems, oscillations on very small scales may be energetically convenient, see [3, 4]. Correspondingly, the variational problem  $\int_{\Omega} (|Dw|^2 - 1)^2 dx$  is not lower semicontinuous, and - depending on the boundary data and forcing - does not have minimizers. However, the curvature term  $\sigma^2 |D^2w|^2$  penalizes oscillations on an exceedingly fine scale and thereby ensures existence of minimizers. The solutions then have oscillations on an intermediate scale, which is determined by the competition between the two terms. The analysis of the specific functional proposed by Ortiz and Gioia is reviewed in Sect. 2 below.

The approach of Ortiz an Gioia was later extended to the full vectorial Föppl-von Kármán energy, and also to three-dimensional elasticity. These refinements explained the appearance of oscillations on two different length scales, with coarse oscillations in a direction normal to the boundary, and fine oscillations in the direction tangential to the boundary, as discussed in Sect. 3 below.

Recently interest has been directed to controlling the microstructures by designing the geometry of the debonded region appropriately [5, 6]. The key idea is to introduce a sacrificial layer between the film and the substrate, and then to selectively etch away a part of it, so that the boundary of the debonded region is straight. The film then partially rebonds to the surface, leading to complex patterns of tunnels. A study of these patterns within the Ortiz-Gioia framework, with a variational functional containing the Föppl-von Kármán energy and a fracture term proportional to the debonded area, is presented in Sect. 4. The mathematical analysis leading to the upper bounds of Theorem 6 suggests the presence of different types of patterns in different parameter ranges. The picture is rather easy in the two extreme cases in which the bonding energy per unit area is very small or very large. Indeed, in the first one the patterns observed for completely debonded films give the optimal energy scaling, in the second one the optimal state corresponds to the film completely bound to the substrate. In the intermediate regime we expect a richer picture, with bonded areas separated by thin debonded tunnels. For a certain regime, depending on the relation between the bonding energy per unit area, the film thickness and the compression ratio, a construction in which the tunnels branch and refine close to the boundary has a lower energy than the one with straight tunnels, see discussion in Sect. 4 below. The microstructure formation in thin films can be understood at a qualitative level as a form of Euler buckling instability. The relevant experiments, however, are well beyond the stability threshold, as discussed in Sect. 5 below.

#### 2 Scalar Modeling of Compressed Thin Films

Ortiz and Gioia showed that, if tangential displacements are neglected, the energy of a compressed thin film can be characterized by the functional

$$I_{\sigma}[w] = \int_{\Omega} \left( \left( |Dw|^2 - 1 \right)^2 + \sigma^2 |D^2w|^2 \right) dx,$$
(1)

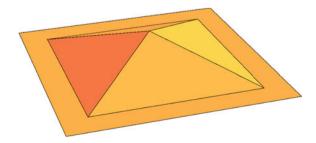
subject to w = 0 on  $\partial \Omega$  and  $w \ge 0$  in  $\Omega$ . Here  $\Omega \subset \mathbb{R}^2$  represents the debonded region,  $w : \Omega \to [0, \infty)$  the rescaled normal displacement, and  $\sigma$  is a small parameter related to the thickness of the film. This functional arises also naturally in the study of liquid crystal configurations [7] and of magnetic structures in thin films [8]. Despite a large mathematical effort [7, 9–15] the problem (1) is not yet completely understood; it has been shown that the minimal energy is proportional to  $\sigma$  but the  $\Gamma$ -limit of  $\sigma^{-1}I_{\sigma}[w]$  has only been partially identified. The natural candidate is

$$I_0[w] = \frac{1}{3} \int_{J_{Dw}} |[Dw]|^3 d\mathcal{H}^1$$
(2)

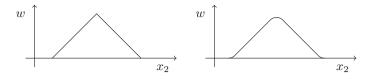
restricted to functions  $w : \Omega \to \mathbb{R}$  which solve the Eikonal equation |Dw| = 1 and are sufficiently regular. Here,  $J_{Dw}$  denotes the set of points (typically, a curve) where the gradient Dw is not continuous, [Dw] denotes its jump across the interface, and  $d\mathcal{H}^1$  the line integral along the interface. In particular, under the additional assumption that Dw is a function of bounded variation, it has been shown that for  $\sigma \to 0$  the scaled functionals  $\sigma^{-1}I_{\sigma}$  converge, in the sense of  $\Gamma$ -convergence, to  $I_0$ , see [11–13] for the lower bound and [14–16] for the upper bound. However, it is also clear that finiteness of the energy does not imply that Dw has bounded variation, but only that w belongs to a larger space, called  $AG(\Omega)$ , see [10, 11]. Therefore the result is still incomplete.

The Eikonal equation |Dw| = 1 with the boundary data w = 0 on  $\partial \Omega$  is solved by the distance to the boundary,  $w_0(x) = \text{dist}(x, \partial \Omega)$ . For example, if  $\Omega$  is a square this leads to the tent-shaped deformation illustrated in Fig. 1. The function  $w_0$  is however only Lipschitz continuous, not twice differentiable, and makes the curvature term

Fig. 1 Sketch of a deformation achieving the optimal energy in (1). Here the debonded region  $\Omega = (0, 1)^2$  is a *square*, and the distance to the boundary gives a "tent"-form. The convolution in (3) makes the folds have smooth transitions on a small scale



D. Bourne et al.



**Fig. 2** Sketch of the effect of the mollification in (3) in a direction orthogonal to the fold. *Left panel*: the distance from the boundary dist(x,  $\partial \Omega$ ) is a function with slope  $\pm 1$  and sharp kinks. *Right panel*: the mollification defined in (3) still has slope  $\pm 1$  on large parts of the domain, but has smooth transitions from one value to the other over a length of the order of  $\sigma$ , see Fig. 3

 $\int_{\Omega} \sigma^2 |D^2 w|^2 dx$  infinite. Therefore Ortiz and Gioia [1, 2] proposed to use a smoothed version of the distance function,

$$w_{\sigma}(x) = \int_{\Omega} \operatorname{dist}(y, \partial \Omega) \varphi_{\sigma}(x - y) dy$$
(3)

where  $\varphi_{\sigma}$  is a mollifier on the scale  $\sigma$ , i.e.,  $\varphi_{\sigma} \in C_{c}^{\infty}(B_{\sigma})$  with  $\int_{\mathbb{R}^{2}} \varphi_{\sigma} dx = 1$  and  $|D\varphi_{\sigma}| \leq c/\sigma^{3}$ . Then the regularized gradient  $Dw_{\sigma}$  has length close to 1 on most of the domain  $\Omega$ , but at boundaries between regions where  $Dw_{0}$  has different orientations  $Dw_{\sigma}$  changes smoothly over a length scale  $\sigma$  from one value to the other. The bending energy is correspondingly localized in a stripe of thickness  $2\sigma$  around the interfaces, see Fig. 2. The prediction that minimizers of (1) are well represented by  $w_{\sigma}$  is in good agreement, at least for some geometries, with experimental observations [1, 2].

The work of Ortiz and Gioia was then extended to related problems, showing for example that under anisotropic compression branching-type microstructures appear close to the boundary [17, 18], or that in certain regimes telephone-cord blisters develop [19–21] thanks to the interaction between the elastic deformation and the fracture problem that determines the boundary of the debonded region.

#### **3** Pattern Formation in Debonded Thin Films

A finer analysis of the nonlinear elasticity model that had led to (1) showed that, in the case of isotropic compression, also the in-plane components exhibit fine-scale oscillations which refine close to the boundary [22–25]. This analysis was based on the Föppl-von Kármán model, which includes the tangential components of the displacement *u* as well. After rescaling the energy takes the form (in the case of zero Poisson's ratio for simplicity)

$$E_{\sigma}[u,w] = \int_{\Omega} \left( |Du + Du^{T} + Dw \otimes Dw - \mathrm{Id}|^{2} + \sigma^{2} |D^{2}w|^{2} \right) dx.$$
(4)

Here  $\Omega \subset \mathbb{R}^2$  is, as above, the debonded region, and the displacements u and w vanish at the boundary of  $\Omega$ , corresponding to the fact that the rest of the film is still bound to the substrate. The isotropic compressive strain has been scaled to 1, and one can check that  $E_{\sigma}[0, w] = 1 + I_{\sigma}[w]$ . The key result from [22, 23] was that the minimum energy scales proportional to  $\sigma$ :

**Theorem 1** (From [22, 23]) Let  $\Omega \subset \mathbb{R}^2$  be a bounded domain with piecewise smooth boundary. Then there are two constants  $c_L, c_U > 0$  such that

$$c_L \sigma \le \min\{E_{\sigma}[u, w] : u = 0, w = 0 \text{ on } \partial \Omega\} \le c_U \sigma.$$
(5)

The argument used for proving the lower bound also proves that a finite fraction of the energy is localized in a thin strip close to the boundary.

Similar statements hold if the plate theory in (4) is replaced by a fully threedimensional nonlinear elastic model. For  $v : \Omega \times (0, h) \to \mathbb{R}^3$ , h > 0, we define

$$E_h^{3D}[v] = \frac{1}{h} \int_{\Omega \times (0,h)} W(Dv) dx \tag{6}$$

where  $W : \mathbb{R}^{3\times3} \to [0, \infty)$  is the elastic stored energy density, which vanishes on the set of proper rotations SO(3) and has quadratic growth, in the sense that

$$c\operatorname{dist}^{2}(F, \operatorname{SO}(3)) \le W(F) \le c'\operatorname{dist}^{2}(F, \operatorname{SO}(3))$$
(7)

for some positive constants c and c'. The factor 1/h is included explicitly in (6) to obtain an energy per unit thickness, corresponding to (4).

In the nonlinear case the thickness h of the film and the compression  $\delta$  enter the problem separately, however to leading order and after scaling the optimal energy only depends on the combination  $\sigma = h/\delta^{1/2}$ . In order to understand this expression it is instructive to recall the relation between the three-dimensional problem  $E_h^{3D}$  and its two-dimensional counterpart  $E_{\sigma}$ . In particular, a given pair (u, w) in (4) corresponds to a three-dimensional deformation  $v_{\delta}$  of the form

$$v_{\delta}(x_1, x_2, x_3) = (1 - \delta) \left[ \psi(x_1, x_2) + x_3 n(x_1, x_2) \right]$$
(8)

where

$$\psi(x_1, x_2) = \begin{pmatrix} x_1 + 2\delta u_1(x_1, x_2) \\ x_2 + 2\delta u_2(x_1, x_2) \\ (2\delta)^{1/2} w(x_1, x_2) \end{pmatrix}$$
(9)

represents the deformation of the  $x_3 = 0$  layer and

$$n(x_1, x_2) = \begin{pmatrix} -(2\delta)^{1/2} \partial_1 w(x_1, x_2) \\ -(2\delta)^{1/2} \partial_2 w(x_1, x_2) \\ 1 \end{pmatrix}$$
(10)

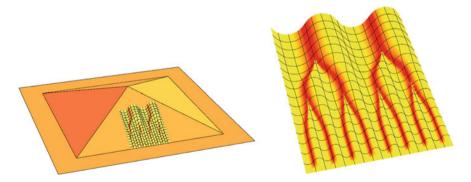
is, to leading order, the normal to the surface described by  $\psi$  and gives the out-ofplane component of the strain. An expansion of  $E_h^{3D}[v_\delta]$  for small  $\delta$  shows that the leading order contribution is proportional to  $\delta^2 E_\sigma[u, w]$  if the Poisson's ratio of the material vanishes. See for example [22, App. A and App. B] for a more detailed discussion of this point. A rigorous relation between  $E_\sigma$  and  $E_h^{3D}$  was derived in [26, 27] by means of  $\Gamma$ -convergence, these results however are appropriate for a different regime, with much smaller energy, and therefore do not apply directly to the situation of interest here.

**Theorem 2** (From [25]) Let  $\Omega \subset \mathbb{R}^2$  be a bounded domain with piecewise smooth boundary,  $\delta \in (0, 1)$ ,  $h \in (0, \delta^{1/2})$ . Then there are two constants  $c_L, c_U > 0$  such that

$$c_L \sigma \le \min\{\frac{1}{\delta^2} E_h^{3D}[u] : u(x) = (1 - \delta)x \text{ for } (x_1, x_2) \in \partial \Omega\} \le c_U \sigma$$
(11)

where  $\sigma = h/\delta^{1/2}$ .

The significance of Theorems 1 and 2 is best understood by considering the key ideas in the proofs. The upper bound in (5) and (11) is proven by explicitly constructing a suitable deformation field (u, w). This is done in several steps. The first step is the Ortiz-Gioia construction given in (3), which correctly describes the large-scale behavior of the film and relaxes the compression in direction normal to the boundary, as in Fig. 1. In the second step one adds fine-scale oscillations in the orthogonal direction, as illustrated in Fig. 3. This microstructure does not change the average shape significantly but relaxes the strain component tangential to the boundary. Finally, one realizes that optimal deformations have oscillations on a very fine scale close to the boundary, to adequately match the boundary data, but much coarser oscillations in



**Fig. 3** Sketch of a deformation achieving the optimal upper bound in (5). As in Fig. 1, the debonded region  $\Omega = (0, 1)^2$  is a square. The starting point, at a coarse scale, is the "tent"-form illustrated in Fig. 1. At a finer scale, folds orthogonal to the boundary relax the tangential compression (*left panel*, folds are only drawn in a small region). The period of the folds is of order *h* close to the boundary, and via a sequence of period-doubling steps becomes coarser in the inside (*right*, blow-up of the folds from the *left panel*)

the interior, to minimize the bending energy. Therefore a number of period-doubling steps are inserted, as illustrated in Fig. 3. Analogous self-similar branched patterns had previously appeared in the study of microstructures in shape-memory alloys [28, 29], where for a simplified model it had been possible to show that minimizers are indeed asymptotically self-similar [30]. The scaling in the presence of finite elasticity, both of the martensite and in the surrounding austenite, was then studied in [31, 32]; vectorial variants of the model were considered in [33, 34]. A similar approach has been useful also for a variety of other problems, ranging from magnetic patterns in ferromagnets [35–37] to field penetration in superconductors [38, 39], dislocation structures in crystal plasticity [40] and coarsening in thin film growth [41].

This variational approach to microstructure formation in thin elastic sheets is much more general, and indeed it can be applied to a number of related problems. One example is paper crumpling [42, 43] in which a thin plate, completely detached from the substrate, is confined to a small volume. In this case it has been possible to construct deformations with much smaller energy per unit volume. In particular one can obtain an energy per unit thickness proportional to  $h^{5/3}$  [44, 45], and one can approximate any compressive deformation with this energy.

**Theorem 3** (From [45]) Let  $\Omega \subset \mathbb{R}^2$  be a bounded domain, r > 0. Then there is a map  $v : \Omega \times (0, h) \to B_r(0)$  such that

$$E_h^{3D}[v] \le ch^{5/3} \,. \tag{12}$$

The constant c may depend on  $\Omega$  and r but not on h. Further, if  $v_0 : \Omega \to \mathbb{R}^3$  is a short map, i.e., a map which obeys  $|v_0(x) - v_0(y)| \le |x - y|$  for all  $x, y \in \Omega$ , then there is a sequence  $v_h$ , converging to  $v_0$ , such that

$$\lim_{h \to 0} \frac{1}{h^{\alpha}} E_h^{3D}[v_h] = 0 \tag{13}$$

for any  $\alpha < 5/3$ . Convergence of  $v_h$  is understood as uniform convergence of the vertical averages.

The proof of this is based on the combination of three ingredients. The first one is an approximation of short maps with Origami maps:

**Theorem 4** (From [45]) Let  $v_0 : \Omega \to \mathbb{R}^3$  be a short map, i.e., a map which obeys  $|v_0(x) - v_0(y)| \le |x - y|$  for all  $x, y \in \Omega$ . Then there is a sequence  $v_j$  of Origami maps converging uniformly to  $v_0$ .

Here we say that a map  $v : \mathbb{R}^2 \to \mathbb{R}^3$  is an Origami map if it is continuous and piecewise isometric, i.e., if the domain can be subdivided into pieces such that v is a linear isometry (a translation plus a rotation) in each piece. The number of pieces is allowed to diverge only at infinity, in the sense that only finitely many pieces are allowed in any bounded subset of  $\mathbb{R}^2$ .

The second step is to approximate any Origami maps with low-energy maps:

**Theorem 5** (From [45]) Let  $\Omega \subset \mathbb{R}^2$  be a bounded domain,  $v_0 : \Omega \times (0, h) \rightarrow B_r(0)$  be an Origami map. Then for any Origami map  $v_0$  there is a sequence of maps  $v_h : \Omega \times (0, h) \rightarrow \mathbb{R}^3$ , converging to  $v_0$ , such that

$$E_h^{3D}[v_h] \le Ch^{5/3} \,. \tag{14}$$

*The constant may depend on*  $\Omega$  *and*  $v_0$  *but not on* h*.* 

This is proven by an explicit construction around each fold.

Another related problem of high current interest is the study of wrinkling patterns in graphene sheets [46, 47]. This has been addressed by a similar model, in which the boundary conditions are replaced by a viscous term describing the interaction with a substrate [48–50]. It would be interesting to see if the methods discussed here can be useful also for this variant of the problem.

### **4** Pattern Formation in Rebonded Thin Films

The microstructures spontaneously developed by compressed thin films can be controlled if the geometry of the debonded region is designed appropriately [5, 6]. One possibility is to introduce a sacrificial layer between the film and the substrate, and then to selectively etch away a part of it, so that the boundary of the debonded region is straight, see sketch in Fig. 4. The film then partially rebonds to the surface, leading to complex patterns of tunnels, which in some cases refine close to the boundary, see Fig. 5.

These patterns can be studied by coupling the von-Kármán energy with a fracture term proportional to the debonded area,

$$E_{\sigma,\gamma}[u,w] = \int_{\Omega} \left( |Du + Du^T + Dw \otimes Dw - \mathrm{Id}|^2 + \sigma^2 |D^2w|^2 \right) dx + \gamma |\{w > 0\}|.$$
(15)

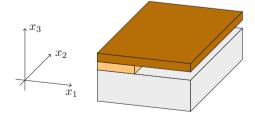
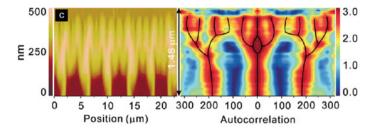


Fig. 4 Geometry of the partially delaminated film. The intermediate sacrificial layer is removed chemically only for  $x_1 > 0$ . The free-standing film is subject to compression at the Dirichlet boundary  $x_1 = 0$  and may rebond to the substrate



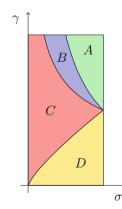
**Fig. 5** Experimental picture of tube branching in  $Si_{1-x}Ge_x$  film on a thick SiO<sub>2</sub> substrate. *Left* AFM image of the network near the etching front. *Right* autocorrelation pattern. Reprinted from [5, Fig. 2] with permission from Wiley

The three terms represent stretching, bending and bonding energies respectively. Here  $u : \Omega \to \mathbb{R}^2$  are the (scaled) tangential displacements and  $\gamma > 0$  is the bonding energy per unit area (related to Griffith's fracture energy),  $|\{w > 0\}|$  represents the area of the set where the vertical displacement *w* is nonzero. Equivalently one could take the debonded state as reference and consider a negative term proportional to the rebonded area,  $-\gamma |\{w = 0\}|$ ; the two energies only differ by an additive constant. The appropriate boundary conditions correspond to the film being bound to a substrate on one side of the domain; for simplicity we shall focus on  $\Omega = (0, 1)^2$  with u = 0 and w = 0 on the  $\{x_1 = 0\}$  side of  $\Omega$ . As above, we assume  $w \ge 0$  everywhere.

The mathematical analysis of the energy (15) leads to the rich phase diagram sketched in Fig. 6, which contains four different regimes [51] that we now illustrate.

For large specific bonding energy  $\gamma$  the film is completely bound to the substrate. In particular the film is flat, so that there is no bending energy, but the stretching energy is not released. The total energy is then proportional to the area of  $\Omega$ , and one obtains  $E_{\sigma,\gamma}[0, 0] = 2$ . This is regime A in Fig. 6 and Theorem 6.

The opposite case of very small bonding energy  $\gamma$  is also easy to understand after the foregoing discussion: here the bonding term plays no significant role and



**Fig. 6** Phase diagram for  $E_{\sigma,\gamma}[u, w]$  in the  $(\sigma, \gamma)$  plane

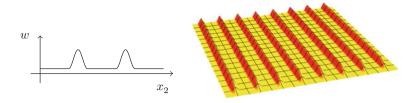


Fig. 7 Sketch of the laminate regime (B)

the film is completely detached from the substrate. One recovers the result of the blistering problem of Theorem 1,  $E_{\sigma,\gamma}[u, w] \simeq E_{\sigma}[u, w] \le c\sigma$ . The corresponding deformations are those illustrated in Fig. 3. This is regime D in Fig. 6 and Theorem 6.

For intermediate values of  $\gamma$  the situation is more complex, in particular debonded channels are formed, which separate wider bonded regions. In regime B the pattern is periodic and, away from the Dirichlet boundary, depends only on the tangential variable  $x_2$ . A large part of the film is bonded to the substrate, but bonded regions are separated by thin tubes, see Fig. 7. Denoting by *h* the period of the oscillations, and by  $\delta$  the width of a tube, the total volume fraction of the tubes is  $\delta/h$ , therefore the bonding energy is proportional to  $\gamma \delta/h$ . Each tube has to release a compression of *h* over a width  $\delta$ , therefore the term  $|Dw|^2$  is of order  $h/\delta$  inside the tubes (the stretching energy is then completely relaxed). This gives  $|Dw| \sim (h/\delta)^{1/2}$  in the tubes, and hence  $|D^2w| \sim (h/\delta)^{1/2}/\delta$ . Therefore the total energy can be estimated by

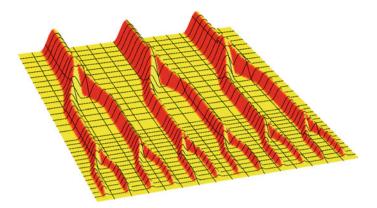
$$\gamma \frac{\delta}{h} + \sigma^2 \frac{\delta}{h} \left( \frac{h^{1/2} / \delta^{1/2}}{\delta} \right)^2 = \gamma \frac{\delta}{h} + \frac{\sigma^2}{\delta^2} \,. \tag{16}$$

Optimizing in  $\delta$  we obtain  $\delta \sim \sigma^{2/3} h^{1/3} \gamma^{-1/3}$  (this is clearly only admissible if  $\delta \leq h \leq 1$ ). The period *h* is fixed by the energetic cost of the interpolation region close to the boundary. In this part of the domain there is no stretch-free construction, and indeed an interpolation over a boundary layer of thickness  $\varepsilon$  results in a total stretching energy of  $\varepsilon(1 + h^2/\varepsilon^2)$ . Optimizing over  $\varepsilon$  we obtain  $\varepsilon \sim h$ , and therefore the total energy for the laminate construction is

$$h + \gamma \frac{\delta}{h} + \frac{\sigma^2}{\delta^2}.$$
 (17)

Inserting the value of  $\delta$  obtained above and minimizing in *h* we conclude that *h* and *E* are proportional to  $(\sigma \gamma)^{2/5}$ . The width of each tube  $\delta$  is then proportional to  $\sigma^{4/5}\gamma^{-1/5}$ . This is regime B in Fig. 6; a precise version of this construction proves the second bound in Theorem 6.

If the bending term becomes more important, it is convenient to insert perioddoubling steps, just like in the discussion of the functional (4). The resulting pattern is shown in Fig. 8. In comparison to the pattern of Fig. 3 the key difference is that the bending is localized to a small region, whereas large parts of the film are bond to the



**Fig. 8** Sketch of the tube branching regime (C)

substrate. The period-doubling steps are only possible at the expense of stretching energy; balancing the different terms one finds [51] that the resulting energy is proportional to  $\sigma^{1/2}\gamma^{5/8}$ . The result of the construction is summarized in the following statement.

**Theorem 6** (From [51]) Let  $\gamma > 0, \sigma \in (0, 1)$ . There are u, w which obey the stated boundary conditions and

$$E_{\sigma,\gamma}[u,w] \le c \begin{cases} 1 & \text{if } \sigma\gamma > 1 & (\text{regime } A), \\ (\sigma\gamma)^{2/5} & \text{if } \sigma^{-4/9} \le \gamma \le \sigma^{-1} & (\text{regime } B), \\ \sigma^{1/2}\gamma^{5/8} & \text{if } \sigma^{4/5} \le \gamma \le \sigma^{-4/9} & (\text{regime } C), \\ \sigma & \text{if } \gamma < \sigma^{4/5} & (\text{regime } D). \end{cases}$$
(18)

The proof is based on making the constructions sketched above precise, details are given in [22] for regime D and in [51] for regimes B and C. Regime A, as discussed above, is immediate.

Optimality of the phase diagram just discussed can be at least partially proven by providing matching lower bounds on the energy. In particular, one can show the following.

**Theorem 7** (From [51]) Let  $\gamma > 0$ ,  $\sigma \in (0, 1)$ . For any u, w which obey the stated boundary conditions one has

$$E_{\gamma,\sigma}[u,w] \ge c \begin{cases} 1 & \text{if } \sigma\gamma > 1 & (\text{regime } A), \\ (\sigma\gamma)^{2/3} & \text{if } \sigma^{1/2} \le \gamma \le \sigma^{-1} & (\text{regime } B'), \\ \sigma & \text{if } \gamma < \sigma^{1/2} & (\text{regime } D'). \end{cases}$$
(19)

Whereas the statement in regime D' follows from [22], the other two bounds are proven in [51] using the Korn-Poincaré inequality for  $SBD^2$  functions obtained in [52].

Theorem 7 proves optimality in phases A and D. The bound in the intermediate region does not, however, match the upper bounds stated in Theorem 6. Therefore it is at this stage not clear if the branching patterns illustrated in Fig. 8 are optimal.

## 5 Linear Stability Analysis

The general form of the linearized Föppl-von Kármán plate theory under isotropic compression is [53, 54]

$$E_{\rm FvK}[u,w] = \frac{1}{2}Yh \int_{\Omega} \left[ (1-\nu)|\varepsilon|^2 + \nu({\rm Tr}\varepsilon)^2 + \frac{h^2}{12} \left[ (1-\nu)|D^2w|^2 + \nu(\Delta w)^2 \right] \right] dx ,$$
(20)

see also [2, 22] for a discussion in the present context and [27] for a rigorous mathematical derivation. Here  $v \in [-1, 1/2]$  is the Poisson ratio, *Y* Young's modulus, *h* the film thickness, and the strain  $\varepsilon$  is defined by

$$\varepsilon = Du + (Du)^T + Dw \otimes Dw - 2\delta \mathrm{Id}, \qquad (21)$$

where  $\delta$  is the eigenstrain (i.e., the compression enforced by the substrate). We recall that we use  $|M|^2 = \text{Tr}M^T M$  for the matrix norm. For  $\nu = 0$ , after a rescaling (20) reduces to (4). We recall that in [22, App. B] it was shown that the scaling behavior of the functional  $E_{\text{FvK}}$  is the same for all  $\nu \in (-1, 1/2]$ , hence our results hold also for generic values of the Poisson ratio. Of course, the regime  $\nu \ge 0$  is the most relevant.

For small  $\delta$  one can linearize around the state u = 0, w = 0. After straightforward computations this leads to

$$E_{\rm FvK}^{\rm lin}[u,w] = \frac{1}{2}Yh \int_{\Omega} \left[ (1-\nu)|Du + Du^T - 2\delta \mathrm{Id}|^2 - 4(1-\nu)\delta |Dw|^2 + \nu(2\mathrm{div}u - 4\delta)^2 - 8\delta\nu |Dw|^2 + \frac{h^2}{12} \left[ (1-\nu)|D^2w|^2 + \nu(\Delta w)^2 \right] \right] dx \, .$$

In this linearized functional u and w are decoupled. The dependence on u is convex, hence u = 0 is the minimizer with the given boundary data. The dependence on wis however not necessarily convex. Working for concreteness in a circle of radius R, we can assume w to be radial,  $w(x) = \varphi(|x|)$ , subject to  $\varphi(R) = 0$ , so that

$$Dw(x) = \varphi'(|x|)\frac{x}{|x|}$$

and

$$D^2 w(x) = \varphi''(|x|) \frac{x}{|x|} \otimes \frac{x}{|x|} + \varphi'(|x|) \left(\frac{\operatorname{Id}}{|x|} - \frac{x \otimes x}{|x|^3}\right)$$

Inserting into the energy leads to the one-dimensional variational problem

$$\frac{1}{2}Yh\int_{0}^{R}\left[-4\delta(1+\nu)(\varphi'(r))^{2} + \frac{h^{2}}{12}\left[(\varphi''(r))^{2} + \left(\frac{\varphi'(r)}{r}\right)^{2} + 2\nu\frac{\varphi'(r)\varphi''(r)}{r}\right]\right]rdr.$$

This is positive definite if the first term, of order  $\delta$ , is not larger then the second term, of order  $h^2/R^2$ . Therefore the loss of stability, which corresponds to Euler buckling, occurs at strains  $\delta \sim h^2/R^2$ . Inserting the experimental data from [5], namely,  $h \sim 20$  nm,  $R \sim 10 \,\mu$ m,  $\nu \sim 0.277$ , leads to  $\delta_{\rm crit} \sim 4 \times 10^{-6}$ , which corresponds to a strain of 0.0004 %. This is over three orders of magnitude smaller than the experimentally applied strain  $\delta_{\rm Exp} \sim 0.011 = 1.1$  %. Therefore the experiments we discussed take place well beyond the loss of linear stability, and a buckling-postbuckling analysis does not seem appropriate to understand the deformations. Our variational approach is instead constructed to deal with deformations and microstructures that appear in the deeply nonlinear regime and is therefore more suitable to study the mentioned experiments.

#### References

- 1. Ortiz, M., & Gioia, G. (1994). Dynamically propagating shear bands in impact-loaded prenotched plates II. Numerical simulations. *Journal of the Mechanics and Physics of Solids*, 42, 531.
- Gioia, G., & Ortiz, M. (1997). Delamination of compressed thin films. Advances in Applied Mechanics, 33, 119.
- 3. Dacorogna, B. (1989). Direct methods in the calculus of variations. *Applied Mathematical Sciences*, 78. Springer.
- 4. Müller, S.: F. Bethuel et al. (Ed.) *Calculus of variations and geometric evolution problems*. Lecture Notes in Math (1713, pp. 85–210). Springer.
- Mei, Y., Thurmer, D. J., Cavallo, F., Kiravittaya, S., & Schmidt, O. G. (2007). Semiconductor sub-micro-/nanochannel networks by deterministic layer wrinkling. *Advanced Materials*, 19, 2124.
- 6. Cendula, P., Kiravittaya, S., Mei, Y. F., Deneke, C., & Schmidt, O. G. (2009). Bending and wrinkling as competing relaxation pathways for strained free-hanging films. *Physical Review B*, 79, 085429.
- Aviles, P., & Giga, Y. (1987). A mathematical problem related to the physical theory of liquid crystal configurations. *Proceedings of the Centre for Mathematical Analysis, Australian National University*, 12, 1.
- DeSimone, A., Kohn, R. V., Müller, S., & Otto, F. (2000). Magnetic microstructures : a paradigm of multiscale problems. In J.M. Ball (ed.) *ICIAM 99: proceedings of the Fourth International Congress on Industrial and Applied Mathematics* (pp. 175–190). Oxford: Oxford University Press.
- 9. Aviles, P., & Giga, Y. (1996). The distance function and energy. *Proceedings of the Royal Society of Edinburgh Section A*, *126*, 923.

- Aviles, P., & Giga, Y. (1999). On lower semicontinuity of a defect energy obtained by a singular limit of the Ginzburg-Landau type energy for gradient fields. *Proceedings of the Royal Society* of Edinburgh Section A, 129, 1.
- 11. Ambrosio, L., De Lellis, C., & Mantegazza, C. (1999). Line energies for gradient vector fields in the plane. *Calculus of Variations and Partial Differential Equations*, 9, 327.
- 12. Jin, W., Kohn, R.V. (2000). Singular perturbation and the energy of folds. *Journal of Nonlinear Science*, 10, 355.
- DeSimone, A., Kohn, R. V., Müller, S., & Otto, F. (2001). A compactness result in the gradient theory of phase transitions. *Proceedings of the Royal Society of Edinburgh Section A*, 131, 833.
- 14. Poliakovsky, A. (2007). Upper bounds for singular perturbation problems involving gradient fields. *Journal of the European Mathematical Society*, 9(1), 1.
- 15. Conti, S., & De Lellis, C. (2007). Sharp upper bounds for a variational problem with singular perturbation. *Mathematische Annalen*, *338*, 119.
- 16. Poliakovsky, A. (2005). A method for establishing upper bounds for singular perturbation problems. *Comptes Rendus Mathematique Academy of Science Paris*, *341*, 97.
- 17. Gioia, G., DeSimone, A., Ortiz, M., & Cuitino, A. M. (2002). Folding energetics in thin-film diaphragms. *Proceedings of the Royal Society of London. Series A*, *458*, 1223.
- Conti, S., DeSimone, A., & Müller, S. (2005). Self-similar folding patterns and energy scaling in compressed elastic sheets. *Computer Methods in Applied Mechanics and Engineering*, 194, 2534.
- 19. Gioia, G., & Ortiz, M. (1998). Determination of thin-film debonding parameters from telephone-cord measurements. *Acta Materialia*, 46(1), 169.
- Audoly, B. (1999). Stability of straight delamination blisters. *Physical Review Letters*, 83, 4124.
- 21. Audoly, B. (2000). Mode-dependent toughness and the delamination of compressed thin films. *Journal of the Mechanics and Physics of Solids*, 48, 2315.
- Ben Belgacem, H., Conti, S., DeSimone, A., Müller, S. (2000). Rigorous bounds for the Föpplvon Kármán theory of isotropically compressed plates. *Journal of Nonlinear Science*, 10, 661.
- Jin, W., & Sternberg, P. (2001). Energy estimates for the von Kármán model of thin-film blistering. *Journal of Mathematical Physics*, 42, 192.
- 24. Jin, W., & Sternberg, P. (2002). In-plane displacements in thin-film blistering. *Proceedings of the Royal Society of Edinburgh Section A*, 132, 911.
- 25. Ben Belgacem, H., Conti, S., DeSimone, A., Müller, S. (2002). Energy scaling of compressed elastic films. *Archive for Rational Mechanics and Analysis*, *164*, 1.
- Friesecke, G., James, R. D., & Müller, S. (2002). A theorem on geometric rigidity and the derivation of nonlinear plate theory from 3d elasticity. *Communications on Pure and Applied Mathematics*, 55, 1461.
- Friesecke, G., James, R. D., & Müller, S. (2006). A hierarchy of plate models derived from nonlinear elasticity by Gamma-convergence. *Archive for Rational Mechanics and Analysis*, *180*, 183.
- Kohn, R. V., & Müller, S. (1992). Branching of twins near an austenite-twinned-martensite interface. *Philosophical Magazine A*, 66, 697.
- Kohn, R. V., & Müller, S. (1994). Surface energy and microstructure in coherent phase transitions. *Communications on Pure and Applied Mathematics*, 47, 405.
- Conti, S. (2000). Branched microstructures: scaling and asymptotic self-similarity. Communications on Pure and Applied Mathematics, 53, 1448.
- 31. Conti, S. (2006). A lower bound for a variational model for pattern formation in shape-memory alloys. *Continuum Mechanics and Thermodynamics*, 17, 469.
- 32. Zwicknagl, B. (2014). Microstructures in low-hysteresis shape memory alloys: scaling regimes and optimal needle shapes. *Archive for Rational Mechanics and Analysis*, 213, 355.
- 33. Diermeier, J. (2013). Master's thesis, Universität Bonn.
- Chan, A., & Conti, S. (2015). Energy scaling and branched microstructures in a model for shape-memory alloys with SO (2) invariance. *Mathematical Models and Methods in Applied Sciences*, 25, 1091.

- Choksi, R., & Kohn, R. V. (1998). Bounds on the micromagnetic energy of a uniaxial ferromagnet. *Communications on Pure and Applied Mathematics*, 51, 259.
- Choksi, R., Kohn, R. V., & Otto, F. (1999). Domain branching in uniaxial ferromagnets: a scaling law for the minimum energy. *Communications on Pure and Applied Mathematics*, 201, 61.
- 37. Viehmann, T. (2009). Uniaxial ferromagnets. Ph.D. thesis, Universität Bonn.
- 38. Choksi, R., Kohn, R. V., & Otto, F. (2004). Energy minimization and flux domain structure in the intermediate state of a type-I superconductor. *Journal of Nonlinear Science*, *14*, 119.
- Choksi, R., Conti, S., Kohn, R. V., & Otto, F. (2008). Ground state energy scaling laws during the onset and destruction of the intermediate state in a type-I superconductor. *Communications* on Pure and Applied Mathematics, 61, 595.
- 40. Conti, S., & Ortiz, M. (2005). Dislocation microstructures and the effective behavior of single crystals. *Archive for Rational Mechanics and Analysis*, *176*, 103.
- 41. Conti, S., & Ortiz, M. (2008). Minimum principles for the trajectories of systems governed by rate problems. *Journal of the Mechanics and Physics of Solids*, *56*, 1885.
- 42. Lobkovsky, A. E., Gentges, S., Li, H., Morse, D., & Witten, T. A. (1995). Boundary layer analysis of the ridge singularity in a thin plate. *Science*, 270, 1482.
- 43. Witten, T. A. (2007). Stress focusing in elastic sheets. Reviews of Modern Physics, 79, 643.
- 44. Venkataramani, S. C. (2004). Lower bounds for the energy in a crumpled elastic sheet a minimal ridge. *Nonlinearity*, *17*, 301.
- 45. Conti, S., & Maggi, F. (2008). Confining thin elastic sheets and folding paper. Archive for Rational Mechanics and Analysis, 187, 1.
- 46. Li, X., Cai, W., An, J., Kim, S. et al. (2009). Large-area synthesis of high-quality and uniform graphene films on copper foils. *Science*, *324*(5932), 1312.
- 47. Zang, J., Ryu, S., Pugno, N., Wang, Q., Tu, Q., Buehler, M. J., et al. (2013). Multifunctionality and control of the crumpling and unfolding of large-area graphene. *Nature Materials*, *12*, 321.
- Zhang, K., & Arroyo, M. (2013). Adhesion and friction control localized folding in supported graphene. *Journal of Applied Physics*, 113, 193501.
- 49. Zhang, K., & Arroyo, M. (2014). Understanding and strain-engineering wrinkle networks in supported graphene through simulations. *Journal of Mechanics and Physics of Solids*, 72, 61.
- 50. Arroyo, M., Zhang, K., & Rahimi, M. (2014). Mechanics of confined solid and fluid thin films: Graphene and lipid bilayers. *IUTAM Symposium on innovative numerical approaches for materials and structures in multi-field and multi-scale problems.*
- Bourne, D., Conti, S., & Müller, S. (2015). Energy bounds for a compressed elastic film on a substrate. *Preprint* arXiv:1512.07416
- 52. Chambolle, A., Conti, S., & Francfort, G. (2014). Korn-Poincaré inequalities for functions with a small jump set. *To appear in Indiana University Mathematics Journal. Preprint* hal-01091710v1.
- 53. Antman, S. S. (1995). Applied Mathematical Sciences. In *Nonlinear problems in elasticity*, 107. Springer.
- 54. Ciarlet, P. G. (1997). Theory of plates, Mathematical elasticity, vol. II. Elsevier.