

Indentation of suspended two-dimensional solids: The signatures of geometrical and material nonlinearity

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Abstract

The material characterization of ultra-thin solid sheets, including two-dimensional materials like graphene, is often performed through indentation tests on a flake suspended over a hole in a substrate. While this ‘suspended indentation’ is a convenient means of measuring properties such as the stretching (two-dimensional) modulus of such materials, experiments on ostensibly similar systems have reported very different material properties. In this paper, we present a modelling study of this indentation process assuming elastic behaviour. In particular, we investigate the possibility that the reported differences may arise from different geometrical parameters and/or non-Hookean deformations, which lead to the system exploring nonlinearities with geometrical or material origins.

Keywords: Two-dimensional solids, Materials characterization, Elastic constants

81.07.-b, 83.60.-a, 62.20.Dc

1. Introduction

Just as it is natural to test the inflation of a tyre and ripeness of fruit by poking with a finger, a common means of testing the mechanical properties of solids is via indentation tests. While much attention has focussed on the determination of bulk elastic constants via the indentation of a half-space (see Harding and Sneddon, 1945; Nix and Gao, 1998; Perriot and Barthel, 2004; Butt et al., 2005; McKee et al., 2011, for example), a great deal of recent interest has focussed on the use of indentation to determine the mechanical properties of two-dimensional materials like graphene and molybdenum disulphide (see Castellanos-Gomez et al., 2015; Cao and Gao, 2019, for reviews). Flakes of such two-dimensional materials are difficult (if not impossible) to manipulate in a tensile testing machine but may be deposited on a substrate relatively easily. If the substrate on which deposition occurs is patterned with holes, the flake is then ‘suspended’ over the holes: indenting the thin material at a point where it is suspended, for example with an Atomic Force Microscope (AFM), yields a response that is largely independent of the substrate’s mechanical properties (provided that the adhesion between substrate and thin layer is sufficient to guarantee clamping at the hole edge). In particular, indentation yields a force–displacement response that is controlled by the stretching stiffness (or two-dimensional Young’s modulus), E_{2D} , of the material, together with any residual tension or bending stiffness of the suspended material.

From the point of view of mechanics, the indentation of suspended two-dimensional materials has much in common with the indentation of suspended elastic membranes. This problem was studied first by Schwerin (1929), but has since been extended to account for the effects of pre-tension

(Norouzi et al., 2006), bending stiffness (Wan et al., 2003) and indenter geometry (Begley and Mackin, 2004; Komaragiri et al., 2005). Nevertheless, these modifications often entail approximate analyses (for example, by assuming an ansatz for the form of the solution, as in Begley and Mackin, 2004), rather than deriving asymptotic results from the full governing equations.

The interest in the mechanical properties of two-dimensional materials has been sparked because of their high material strength, their novel electronic properties, and the interaction between imposed elastic strain and electronic properties (Vozmediano et al., 2010; Akinwande et al., 2017; Harats et al., 2020); determining the value of the stretching stiffness E_{2D} , as well as any pre-existing tension, is therefore the main focus of indentation experiments on suspended two-dimensional materials.

The first measurement of E_{2D} in graphene was made by Lee et al. (2008), who reported a value $E_{2D} \approx 340$ N/m based on AFM indentation experiments on suspended flakes. While this value is in good agreement with that predicted from first principles (Kudin and Scuseri, 2001), values as low as $E_{2D} \approx 20$ N/m at room temperature (Nicholl et al., 2015) and as high as $E_{2D} \approx 680$ N/m with imposed strain (López-Polín et al., 2017) have been reported. It has also been reported that an optimal number of defects may increase the value of E_{2D} (López-Polín et al., 2015).

There are many complicating factors involved in the indentation of truly two-dimensional materials like graphene including the importance of thermal crumpling (flexural phonons) and static wrinkles that may both give rise to ‘hidden area’ (Nicholl et al., 2017), the anisotropy induced by the underlying hexagonal lattice (Kumar and Parks, 2015) as well as the possibility of slip at the boundary. In particular, excess membrane area that is hidden in static wrinkles and thermal fluctuations may lead to measurements of the stretching modulus E_{2D} that are small and/or load-dependent simply because out-of-plane deformations are ironed out at low applied stress, as is common when extra material is ‘buffered-by-buckling’ (Vella, 2019). This hidden area may have the effect of a strain-dependent stretching modulus and hence be a major cause of the discrepancy in values of E_{2D} reported in the literature, while a stress-dependent stretching modulus of two-dimensional solids might also arise because of finite atomic bond lengths. Nevertheless, it has also been pointed out that some inconsistencies may exist in the way that the predictions of classical elasticity theory are used to interpret experimental data (Vella and Davidovitch, 2017; Jia and Ben Amar, 2020). Here, our aim is to set out clearly the predictions of the standard models of mechanics that are appropriate to thin, isotropic elastic solids as derived asymptotically from the governing equations. We aim to highlight the potential pitfalls that experimental attempts to characterize material properties by indentation may fall into, focussing in particular on the predictions of these models in the parameter regimes that are of most relevance to two-dimensional solids. A key question in this work will be how to separate the effect of geometrical nonlinearities (particularly the effect of indenter geometry) from the effect of material nonlinearities (*i.e.* non-Hookean stress-strain responses). By doing so, we hope that future experiments will be more readily reconciled, and more clearly highlight which experimental results are a consequence of the unique properties of these unusual solids.

The paper is organized as follows. In §2 we present our general modelling approach, together with a simple scaling analysis that highlights the variety of possible behaviours; the regimes in which these different behaviours are expected is summarized in Fig. 2. We then move on to consider in more detail the effect of indenter geometry with a linear stress-strain relation by considering a cylindrical indenter (§3) and a spherical-tipped indenter (§4). In §5 we consider the effect of material nonlinearity (*i.e.* non-Hookean behaviour), before discussing the significance of our results

for the experimental determination of elastic constants in §6 and then summarizing our results and concluding in §7.

2. Modelling approach

2.1. Physical model of indentation

Typical experimental measurements of the mechanical properties of two-dimensional solids by indentation involve a sheet being suspended over a circular hole, of radius R_{out} , on an otherwise planar substrate. While the boundary conditions at the edge of the hole are not generally well-controlled, it is usually assumed that the sheet is perfectly clamped at this boundary (*i.e.* there is no additional radial displacement at the edge as indentation progresses) by the film-substrate adhesion. We shall also assume that the clamping is perfect. When the sheet is deposited it is typically subject to a pre-existing tension, or pre-stress, T_{pre} , which may be caused by the processing or fabrication of the sheet. We shall assume that the pre-tension is uniform and isotropic and, further, that all deformations are axisymmetric.

A quantity of considerable practical interest is the two-dimensional Young's modulus of the sheet, E_{2D} . For a thin, Hookean sheet of thickness t , and Young's modulus E , $E_{2D} = Et$, but we use E_{2D} throughout so that our theory describes equally thin elastic sheets and two-dimensional solids. We assume the sheet has a (two-dimensional) Poisson's ratio ν .

Indentation is typically performed by an Atomic Force Microscope (AFM) tip, which applies the force F required to impose a vertical displacement δ of the central region (measured relative to the clamped edges). We consider two tip shapes in detail: a cylindrical indenter (of radius $R_{\text{in}} < R_{\text{out}}$), which facilitates our analysis, and a spherical tip (of radius of curvature $R_s < R_{\text{out}}$), which is more representative of the tips used experimentally (see López-Polín et al., 2017, for example). We shall see that the shape of the indenting tip can be important for the final interpretation of the force-displacement relationship. A sketch of the typical experimental setup is shown in Fig. 1.

2.2. Scalings and physical arguments

The relationship between the indentation force and depth, $F(\delta)$, holds important information for understanding the elastic properties of thin sheets. To get a first sense of the possible different behaviours of the force-indentation relation, we begin by considering at a scaling level the various energies in the problem.

The work done by the indenter, which scales as $\mathcal{U}_{\text{work}} \sim F\delta$, must be stored predominantly in either the stretching or bending energies of the sheet. To estimate the stretching energy stored within the sheet, we note that the sheet tension has two components: one caused by the pre-tension, T_{pre} , and another caused by the stretching in response to the imposed strain $\varepsilon \sim (\delta/R_{\text{out}})^2$, which is $\Delta T \sim E_{2D} \cdot \varepsilon$. (Note that, for small displacements $\delta \ll R_{\text{out}}$, the strain estimate $\varepsilon \sim (\delta/R_{\text{out}})^2$ can be derived from elementary geometrical considerations.) We therefore write $T \sim T_{\text{pre}} + E_{2D}\varepsilon$ and note that the stretching energy of the sheet must therefore scale according to $\mathcal{U}_s \sim R_{\text{out}}^2 (T_{\text{pre}} + E_{2D}\varepsilon)\varepsilon$. Finally, we note that the bending energy of the sheet scales as $\mathcal{U}_b \sim BR_{\text{out}}^2 \mathcal{K}^2$, where $\mathcal{K} \sim \delta/R_{\text{out}}$ is the typical sheet curvature and B is the bending modulus (for an isotropic, thin, Hookean solid, $B = E_{2D}t^2/12(1 - \nu^2)$), though we use a general bending modulus to account

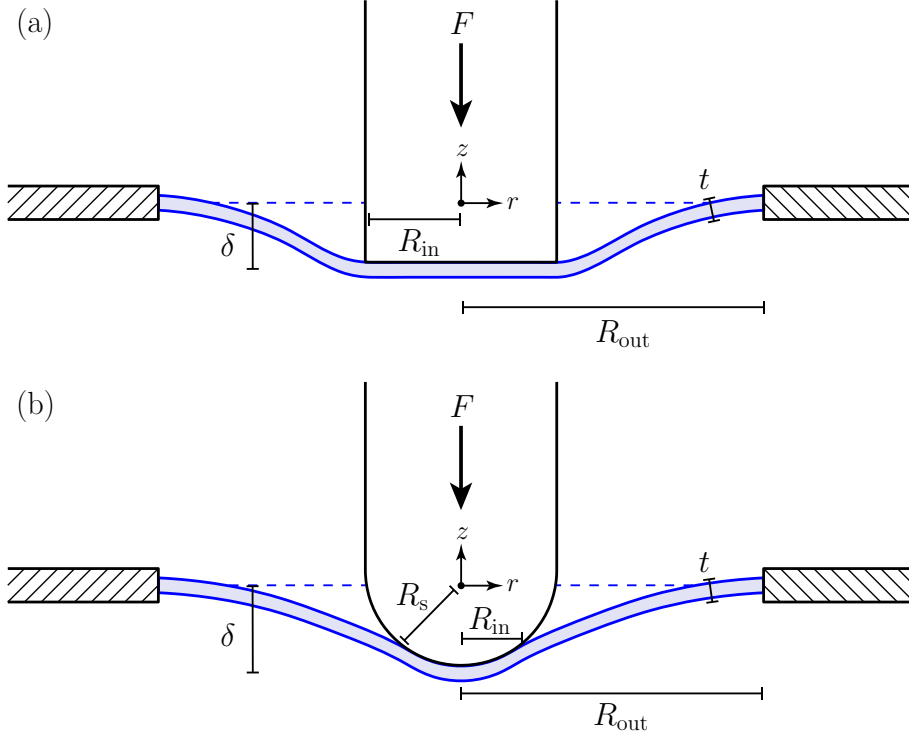


Figure 1: Cross-sectional sketch of the indentation of a clamped sheet by: (a) a cylindrical indenter (or punch) of known radius R_{in} and (b) a spherical-capped indenter of known radius of curvature, R_s , but unknown contact radius, $R_{\text{in}}(\delta)$.

for truly two-dimensional solids, such as graphene, for which the effective value of B may differ significantly from this value¹).

Before considering the different limits, we note that the work done, $\mathcal{U}_{\text{work}}$, must equal the sum of these different energies, *i.e.* $\mathcal{U}_{\text{work}} \sim \mathcal{U}_{\text{b}} + \mathcal{U}_{\text{s}}$, and hence

$$F \cdot \delta \sim \underbrace{R_{\text{out}}^2 T_{\text{pre}} \left(\frac{\delta}{R_{\text{out}}} \right)^2}_{\text{pre-tension}} + \underbrace{R_{\text{out}}^2 E_{2D} \left(\frac{\delta}{R_{\text{out}}} \right)^4}_{\text{stretching}} + \underbrace{R_{\text{out}}^2 B \left(\frac{\delta}{R_{\text{out}}^2} \right)^2}_{\text{bending}}. \quad (1)$$

Written in this way, there are three possible dominant balances, depending on which term dominates the RHS of (1): (i) pre-tension dominated so that $F \sim T_{\text{pre}}\delta$, corresponding to linear membrane theory with constant tension (see Begley and Mackin, 2004; Komaragiri et al., 2005; Vella and Davidovitch, 2017, for example); (ii) stretching dominated so that $F \sim E_{2D}\delta^3/R_{\text{out}}^2$, recovering the scaling of the classical Schwerin solution (see Schwerin, 1929; Begley and Mackin, 2004; Komaragiri et al., 2005; Vella and Davidovitch, 2017); (iii) bending stiffness dominated so

¹Typically the bending modulus of graphene is given as around 1.2 eV (see Lu et al., 2009; Akinwande et al., 2017, for example), or $B \approx 1.9 \times 10^{-19}$ J; however, based on the stretching modulus $E_{2D} = 340$ Nm and thickness $t \approx 0.335$ nm (Akinwande et al., 2017), the corresponding isotropic solid would have $B = E_{2D}t^2/12(1 - \nu^2) \approx 3.4 \times 10^{-18}$ J. As a result, graphene is 10 times more bendable than would be expected for a corresponding isotropic solid; this reflects the fact that, since it is only a single molecule thick, the usual mechanism for generating a bending stiffness (*i.e.* differential strain through the sheet thickness) is not relevant for graphene.

that $F \sim B\delta/R_{\text{out}}^2$, and the sheet responds as a classical plate (see Timoshenko and Woinowsky-Krieger, 1959; Wan et al., 2003; Komaragiri et al., 2005, for example).

Taking the pre-tension dominated case as a reference state, natural choices of dimensionless indentation depth, force, and bending stiffness are

$$\mathcal{F} := \frac{FE_{2D}^{1/2}}{2\pi T_{\text{pre}}^{3/2} R_{\text{out}}}, \quad d := \frac{\delta}{R_{\text{out}}} \left(\frac{E_{2D}}{T_{\text{pre}}} \right)^{1/2}, \quad \mathcal{B} := \frac{B}{T_{\text{pre}} R_{\text{out}}^2}, \quad (2a-c)$$

respectively.

Alternatively, one could have taken the bending or stretching dominated cases as the reference state (Komaragiri et al., 2005, for example). In the applications of current interest (especially for two-dimensional materials) the bending dominated region is of limited interest: for graphene typical values are $B \approx 10^{-19}$ J (Lu et al., 2009) with $T_{\text{pre}} \approx 0.1$ N m⁻¹ and $R_{\text{out}} \approx 1$ μ m (Lee et al., 2008) so that $\mathcal{B} \approx 10^{-6} \ll 1$. The pre-tension therefore dominates the bending stiffness and so we choose a non-dimensionalization that allows the limit $\mathcal{B} \rightarrow 0$ to be easily taken. Similarly, as suggested by Vella and Davidovitch (2017), many recent experiments do not always reach the stretching-dominated (or Schwerin) regime; the non-dimensionalization in (2) allows us to focus on the transition between the dominant balances (i) and (ii).

In dimensionless variables the above dominant balances become: (i) $\mathcal{F} \sim d$ provided $\{\mathcal{F} \ll 1, \mathcal{B} \ll 1\}$, (ii) $\mathcal{F} \sim d^3$ provided $\{\mathcal{F} \gg \mathcal{B}^{3/2}, \mathcal{F} \gg 1\}$, and (iii) $\mathcal{F} \sim \mathcal{B}d$ provided $\{\mathcal{F} \ll \mathcal{B}^{3/2}, \mathcal{B} \gg 1\}$. These behaviours/regions were the main discussion of Komaragiri et al. (2005) who considered indentation by an idealized point-indenter (with our regions (i)–(iii) respectively corresponding to regions 3–1 in Komaragiri et al. 2005). Regions (i)–(iii) are shown in the regime diagrams Fig. 2 along with subregions (associated with the onset of bending and stretching) and asymptotic results — which are both established in Sections 3 & 4. We emphasize that region (i) corresponds to a constant compliance, d/\mathcal{F} , while region (ii) corresponds to a constant ‘cubic compliance’, $d/\mathcal{F}^{1/3}$. (Discussing compliance, d/\mathcal{F} , rather than stiffness, \mathcal{F}/d , simplifies the analytical results presented here.) We shall be focussed in this paper on understanding the dependence of each of these compliances on the material properties of the system, and the behaviour of the system in-between the asymptotic regimes that correspond to regions (i) and (ii).

From these simple energy arguments, one might assume that the indenter’s geometry has little effect on the response $d(\mathcal{F})$. This assumption has been made implicitly across a range of experimental work (Lee et al., 2008; Castellanos-Gomez et al., 2015; López-Polín et al., 2015; López-Polín et al., 2017, to name a few) — in a complex experimental setup, applying the point-indenter ‘solutions’ to experimental data allows progress to be made. In this paper, we investigate the circumstances in which the radius of contact, R_{in} , and the shape of the indenter matter.

For a cylindrical indenter we therefore introduce the dimensionless radius

$$\mathcal{R} := \frac{R_{\text{in}}}{R_{\text{out}}}. \quad (3)$$

For a sphere, it is not immediately clear what length scale should be used to measure the sphere size; here we use the tensile length $R_{\text{out}}(E_{2D}/T_{\text{pre}})^{1/2}$, which is used to rescale vertical deflections; we thus introduce the dimensionless sphere radius:

$$\mathcal{R}_s := \frac{R_s}{R_{\text{out}}} \left(\frac{T_{\text{pre}}}{E_{2D}} \right)^{1/2}. \quad (4)$$

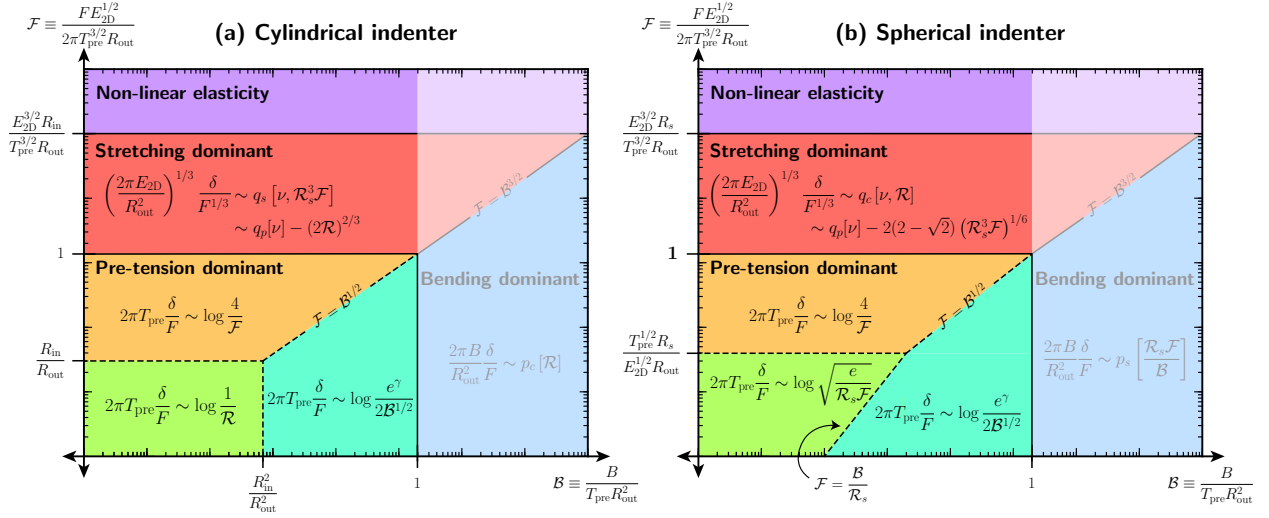


Figure 2: Regime diagram for indentation: shown are the regions of the dimensionless- $(\mathcal{B}, \mathcal{F})$ -space in which different force–displacement relationships are expected to be observed for (a) a cylindrical indenter of dimensionless radius $\mathcal{R} = R_{\text{in}}/R_{\text{out}}$ and (b) a spherical indenter of dimensionless radius $\mathcal{R}_s = T_{\text{pre}}^{1/2} R_s / (E_{2D}^{1/2} R_{\text{out}})$. In this paper, we concentrate only on the effects of small bending stiffnesses $\mathcal{B} \ll 1$.

2.3. Moderate strains and rotations

The energy arguments described in the last section assumed that the stress and strain were linearly related, *i.e.* that the material remains Hookean throughout. This assumption even fails before indentation occurs if the pre-strain $\sim T_{\text{pre}}/E_{2D} = O(1)$ or during indentation if the stretching induced strain $\sim F/(rE_{2D}) = O(1)$ for $R_{\text{in}} \leq r \leq R_{\text{out}}$. In dimensionless variables these two conditions are equivalent to requiring:

$$\mathcal{E} \ll 1 \quad \text{and} \quad \mathcal{F} \ll \frac{\mathcal{R}}{\mathcal{E}^{3/2}}, \quad (5a,b)$$

for a Hookean response, where we have introduced the new dimensionless variable

$$\mathcal{E} := \frac{T_{\text{pre}}}{E_{2D}}, \quad (6)$$

as a measure of the pre-strain in the sheet caused by the pre-tension T_{pre} . (The variable \mathcal{E} is only required for the nonlinear elastic model presented in §5.) In the case of a spherical indenter (for which the radius of contact \mathcal{R} is unknown) an analogous bound to (5b), can be formulated by noting that our detailed analysis in §4 shows that $\mathcal{R} \sim (\mathcal{R}_s^3 \mathcal{F})^{1/4}$, eq. (25b), so that the linear analysis holds provided that $\mathcal{F} \ll \mathcal{R}_s / \mathcal{E}^{3/2}$.

When the strains remain small, it is possible to make reasonable amounts of analytical progress (see Vella and Davidovitch, 2017, for example). However, much of the recent interest in graphene has focussed on whether its material properties change measurably with strain (see *e.g.* Nicholl et al. 2015; López-Polín et al. 2017). We shall, therefore, be interested here in presenting models of indentation in which the material behaviour becomes nonlinear at some point during indentation. In §5 we consider the effects of large strains/slopes and nonlinear constitutive relationships using the theory of finite elasticity. For now, however, we focus on describing the linearly elastic behaviour more fully.

3. Cylindrical indentation of a linearly elastic sheet

We begin by considering the case of a cylindrical indenter, for which the specification of the problem (especially the boundary conditions) is relatively simple.

3.1. Föppl-von Kármán formulation

We initially confine our attention to linear elasticity with the Kirchhoff assumptions (*i.e.* small strains and plate rotations), so that the Föppl-von Kármán (FvK) equations hold (Mansfield, 2005). The axisymmetric FvK equations link the out-of-plane displacement of the sheet, $z(r)$, to the stress via a stress potential $\psi(r)$, which is defined such that the principal stresses are $\sigma_{rr} = \psi(r)/r$ and $\sigma_{\theta\theta} = \psi'(r)$, thereby ensuring that the in-plane equation holds automatically (Mansfield, 2005). The out-of-plane force balance and the compatibility of strains condition may both be integrated once to give:

$$Br \frac{d}{dr} \left[\frac{1}{r} \frac{d}{dr} \left(r \frac{dz}{dr} \right) \right] = \psi \frac{dz}{dr} - \frac{F}{2\pi}, \quad (7a)$$

$$r \frac{d}{dr} \left[\frac{1}{r} \frac{d}{dr} (r\psi) \right] = -\frac{E_{2D}}{2} \left(\frac{dz}{dr} \right)^2, \quad (7b)$$

respectively. Note that, since we are assuming linear elasticity, the deformed and undeformed configurations are interchangeable; the radial coordinate in the sheet is thus denoted $r \in [R_{\text{in}}, R_{\text{out}}]$.

At the outer-rim, $r = R_{\text{out}}$, we assume a perfectly clamped boundary:

$$z(R_{\text{out}}) = 0, \quad z'(R_{\text{out}}) = 0, \quad \psi'(R_{\text{out}}) - \nu \frac{\psi(R_{\text{out}})}{R_{\text{out}}} = (1 - \nu)T_{\text{pre}}. \quad (8a-c)$$

Here (8a,b) are geometric conditions of zero vertical displacement and slope, while (8c) ensures the radial displacement is fixed to be that caused by the initial isotropic tension T_{pre} .

At the inner-rim, $r = R_{\text{in}}$, we assume a perfect-slip boundary (in reality there may be a small amount of adhesion/friction between the indenter and sheet):

$$z(R_{\text{in}}) = -\delta, \quad z'(R_{\text{in}}) = 0, \quad \psi'(R_{\text{in}}) - \frac{\psi(R_{\text{in}})}{R_{\text{in}}} = 0. \quad (9a-c)$$

Here (9a,b) are geometric conditions of continuous vertical displacement and slope; note that (9c) comes from a force balance with the inner (known-geometry) solution [$\psi(r) \propto r$], explaining why (9c) is independent of Poisson ratio ν in contrast to (8c). Throughout this paper we shall assume that the sheet and indenter remain in contact for $r \leq R_{\text{in}}$.

3.1.1. Non-dimensionalization

We use the dimensionless variables suggested by the discussion of energy and scalings in §2.2; in particular, we define

$$\rho := \frac{r}{R_{\text{out}}}, \quad \Psi := \frac{\psi}{T_{\text{pre}} R_{\text{out}}}, \quad Z := \frac{z}{R_{\text{out}}} \left(\frac{E_{2D}}{T_{\text{pre}}} \right)^{1/2}. \quad (10a-c)$$

Substitution of (10) into (7)–(9) gives the dimensionless system

$$\mathcal{B}\rho \frac{d}{d\rho} \left[\frac{1}{\rho} \frac{d}{d\rho} \left(\rho \frac{dZ}{d\rho} \right) \right] = \Psi \frac{dZ}{d\rho} - \mathcal{F}, \quad (11a)$$

$$\rho \frac{d}{d\rho} \left[\frac{1}{\rho} \frac{d}{d\rho} (\rho \Psi) \right] = -\frac{1}{2} \left(\frac{dZ}{d\rho} \right)^2, \quad (11b)$$

for $\mathcal{R} \leq \rho \leq 1$, subject to the boundary conditions

$$Z(1) = 0, \quad Z'(1) = 0, \quad \Psi'(1) - \nu \Psi(1) = 1 - \nu, \quad (12a-c)$$

$$Z(\mathcal{R}) = -d, \quad Z'(\mathcal{R}) = 0, \quad \Psi'(\mathcal{R}) - \frac{\Psi(\mathcal{R})}{\mathcal{R}} = 0. \quad (12d-f)$$

(Recall that \mathcal{F} , \mathcal{B} , and \mathcal{R} are defined in equations (2a), (2c), and (3), respectively.)

For given dimensionless parameters ν , \mathcal{B} , \mathcal{F} , and \mathcal{R} , (11) subject to (12) may be solved by a standard numerical integrator (in our work we used `bvp4c` in `MATLAB`). To make analytical progress we consider separately two asymptotic limits that allow for simplifications: (i) small deflections from the pre-stretched state and (ii) negligible bending stiffness. In §3.2 we consider the first of these, by considering perturbations to the isotropic pre-tensed state ($\psi \sim T_{\text{pre}} r$, $\Psi \sim \rho$), thereby extending the work of *e.g.* Jennings et al. (1995); Wan et al. (2003), while in §3.3 we consider the second case by considering the zero bending stiffness limit ($\mathcal{B} = 0$), similarly to *e.g.* Bhatia and Nachbar (1968); Vella and Davidovitch (2017). This analysis allows us to reproduce some previously known results in a systematic way, whilst also uncovering new results in some regimes; we discuss the broader context of these results as they are derived.

3.2. Small indentation forces ($\mathcal{F} \ll \max\{\mathcal{B}^{1/2}, \mathcal{R}\}$)

For sufficiently small indentations, the stretching of the sheet is negligible compared to the isotropic pre-tension and the (also small) bending stiffness, $\mathcal{B} \ll 1$. To investigate how bending and pre-tension interact, we follow the approach of *e.g.* Vella et al. (2012); Box et al. (2017), and linearize the governing equations (11) about the initial pre-tensed state: we let $\Psi(\rho) \sim \rho + \tilde{\Psi}(\rho)$ and $Z(\rho) \sim 0 + \tilde{Z}(\rho)$ for $\tilde{\Psi}, \tilde{Z} \ll 1$. The two equations (11) thus reduce to a single third-order differential equation for \tilde{Z} that can be solved by a linear combination of logarithms and modified Bessel functions (see Appendix A). Applying the boundary conditions (12) we obtain an explicit relationship between the force and indentation depth that is linear (*i.e.* $\mathcal{F} \propto d$). We express this relationship through the (constant) compliance d/\mathcal{F} :

$$\frac{d}{\mathcal{F}} = \log \frac{1}{\mathcal{R}} + \frac{\hat{K}_1 \hat{I}_0^R + \hat{I}_1 \hat{K}_0^R + \mathcal{R} \left(\hat{K}_0 \hat{I}_1^R + \hat{I}_0 \hat{K}_1^R \right) - 2\mathcal{B}^{1/2} \mathcal{B}^{1/2}}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R} \frac{\mathcal{B}^{1/2}}{\mathcal{R}}, \quad (13)$$

where

$$\hat{I}_j := I_j \left(\mathcal{B}^{-1/2} \right), \quad \hat{I}_j^R := I_j \left(\mathcal{R} \mathcal{B}^{-1/2} \right), \quad (14a,b)$$

$$\hat{K}_j := K_j \left(\mathcal{B}^{-1/2} \right), \quad \hat{K}_j^R := K_j \left(\mathcal{R} \mathcal{B}^{-1/2} \right), \quad (14c,d)$$

and $I_j(x)$ and $K_j(x)$ are the j th-order modified Bessel functions of the first and second kind, respectively (Abramowitz and Stegun, 1964).

Equation (13) corresponds to a constant compliance regime: the compliance d/\mathcal{F} is a function of the indenter radius, \mathcal{R} , and bending stiffness, \mathcal{B} , only. One could use this solution as an explicit formula to describe the small indentation compliance of a tense clamped plate. As a tool for inferring the bending stiffness from an experimental measure of the compliance, however, the complexity of this equation is daunting since the various dimensionless quantities are coupled within the Bessel functions. Instead, recall that we are specifically interested in the limit of small bending stiffnesses $\mathcal{B} \ll 1$ and note, from (14b,d), that $\mathcal{B}/\mathcal{R}^2$ is a key parameter. We therefore consider separately the cases $\mathcal{B} \ll \mathcal{R}^2 < 1$ and $\mathcal{R}^2 \ll \mathcal{B} \ll 1$. We find the leading-order results

$$\frac{d}{\mathcal{F}} \sim \begin{cases} \log \frac{e^\gamma}{2\mathcal{B}^{1/2}} & \text{for } \mathcal{R}^2 \ll \mathcal{B} \ll 1, \\ \log \frac{1}{\mathcal{R}} & \text{for } \mathcal{B} \ll \mathcal{R}^2 < 1, \end{cases} \quad (15)$$

where $\gamma \approx 0.577$ is the Euler–Mascheroni constant (Abramowitz and Stegun, 1964).

The evolution of the compliance with indentation force is shown in Fig. 3 for a fixed \mathcal{R} and four values of \mathcal{B} ; we see that, for small \mathcal{F} , numerical results agree with our asymptotic results. Note, in particular, that the results for $\mathcal{B} = 10^{-8}$ and $\mathcal{B} = 10^{-9}$ are essentially indistinguishable at the scale of the plot, Fig. 3(a): in both cases, $\mathcal{R}^2 = 10^{-6} \gg \mathcal{B}$ and so the small-indentation compliance is controlled by the indenter radius, rather than bending stiffness, as predicted by (15). In these cases, the compliance is essentially indistinguishable from that of an ideal membrane with $\mathcal{B} = 0$.

Overall, for small displacements by a cylindrical indenter ($\mathcal{F} \ll \max\{\mathcal{B}^{1/2}, \mathcal{R}\}$), the indentation compliance d/\mathcal{F} is a constant determined by the relative size of the indenter radius \mathcal{R} and the bending stiffness $\mathcal{B}^{1/2}$. We note also that the fact that the form of the stiffness in each of the cases in (15) is functionally similar — both are logarithmic — is not a coincidence: in the bending-dominated case, the sheet is approximately flat over a region of horizontal scale $\mathcal{B}^{1/2}$, and it is as if the sheet were deformed by a virtual cylindrical indenter of radius $\mathcal{R} \sim 2e^{-\gamma}\mathcal{B}^{1/2}$.

While the indenter-dominated solution (15)[$\mathcal{B} \ll \mathcal{R}^2 < 1$] has been previously derived (see Jennings et al., 1995, for example), we believe that the bending-dominated solution (15)[$\mathcal{R}^2 \ll \mathcal{B} \ll 1$] is novel; this describes scenarios when the bending stiffness is small, but cannot be neglected because the indenter size is smaller than the virtual bending-induced indenter radius, which is therefore the relevant length scale.

3.3. Small bending stiffnesses ($\mathcal{B}^{1/2} \ll \max\{\mathcal{F}, \mathcal{R}\}$)

In many experimental setups the dimensionless bending stiffness \mathcal{B} is small enough that the sheet can be modelled as a thin membrane. In this case, we can simplify the model by taking $\mathcal{B} \rightarrow 0$ in (11a) and dropping the highest-order boundary conditions (12b,e) — ignoring the effects of the edge boundary layers, which occur over a typical length scale $\rho \sim \mathcal{B}^{1/2}$.

Vella and Davidovitch (2017, App. B) solved the membrane problem for a point indenter ($\mathcal{R} = 0$); we extend their work by solving the finite cylinder case (the full derivation can be found in our Appendix B). Ultimately, we obtain the parametric force–displacement relation:

$$d = 2 \frac{\sinh^{-1} \Phi_1^{1/2} - \sinh^{-1} \Phi_0^{1/2}}{A[\Phi_0, \Phi_1; \mathcal{R}, \nu]^{1/2}}, \quad \mathcal{F} = \frac{2}{\mathcal{R}^2} \frac{\Phi_0(1 + \Phi_0^{-1})^{-1/2}}{A[\Phi_0, \Phi_1; \mathcal{R}, \nu]^{3/2}}, \quad (16a,b)$$

$$A[\Phi_0, \Phi_1; \mathcal{R}, \nu] := \frac{2}{1 - \nu} \frac{\Phi_0}{\mathcal{R}^2} \sqrt{\frac{1 + \Phi_1^{-1}}{1 + \Phi_0^{-1}}} - \frac{1 + \nu}{1 - \nu} \Phi_1, \quad (16c)$$

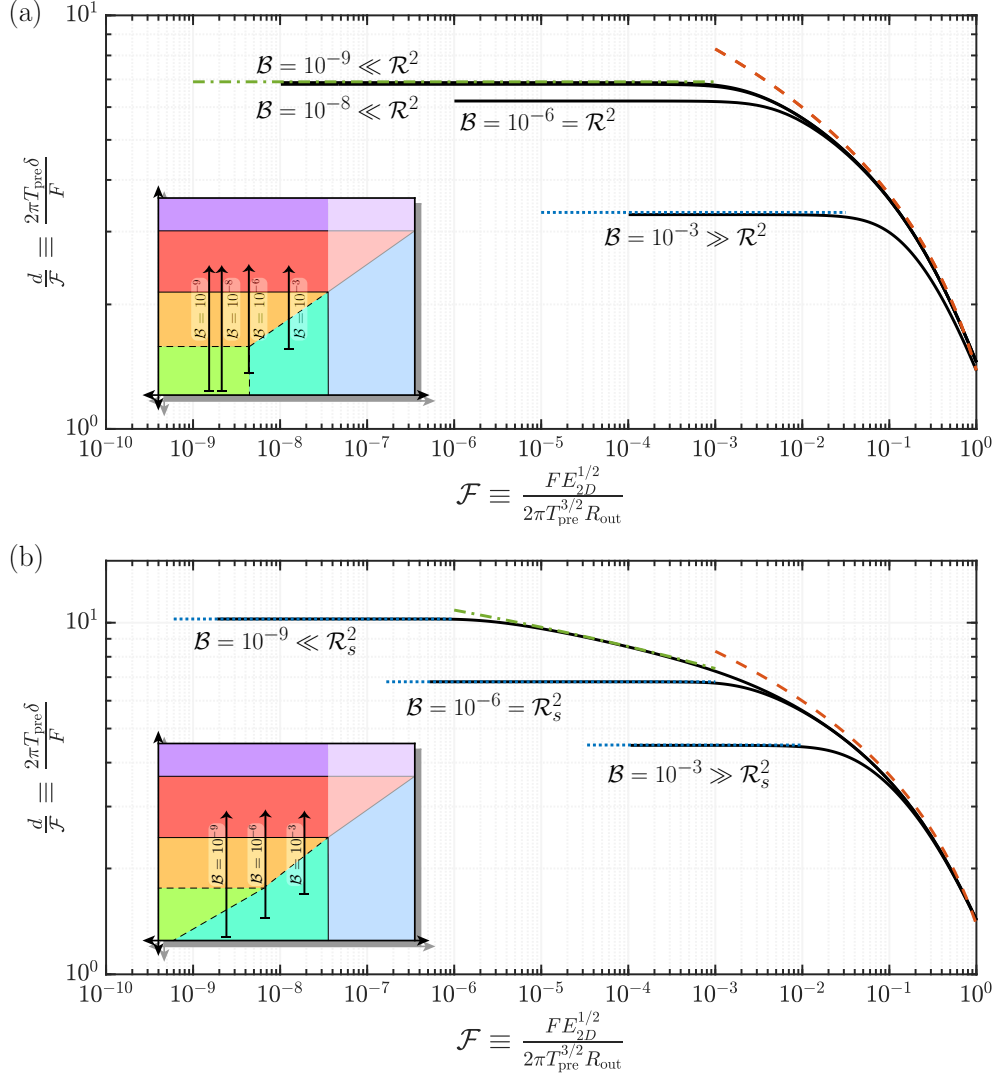


Figure 3: The evolution of the linear compliance as a function of imposed force \mathcal{F} for sheets with different bending stiffnesses ($\mathcal{B} = 10^{-3}, 10^{-6}, 10^{-8}, 10^{-9}$) and $\nu = 1/3$. Results are shown for (a) a cylindrical indenter ($\mathcal{R} = 10^{-3}$) and (b) a spherical indenter ($\mathcal{R}_s = 10^{-3}$). Numerical solutions to the FvK equations are shown as black solid curves while the corresponding asymptotic solutions are shown as broken curves as follows: eqs. (15)[$\mathcal{R}^2 \ll \mathcal{B}$] & (23a)[$\mathcal{R}_s \mathcal{F} \ll \mathcal{B}$] in blue (dotted), eqs. (18)[$\mathcal{F} \ll \mathcal{R}$] & (23a)[$\mathcal{B} \ll \mathcal{R}_s \mathcal{F}$] in green (dash-dotted), and eqs. (18)[$\mathcal{R} \ll \mathcal{F}$] & (24a)[$\mathcal{R}_s \ll \mathcal{F}$] in red (dashed). The insets show the routes through the relevant $(\mathcal{B}, \mathcal{F})$ parameter space (see the regime diagrams of Fig. 2) taken by indentation in each case.

which is parametrized by the boundary stresses $\Phi_1 := A\Psi(1)$ and $\Phi_0 := A\mathcal{R}\Psi(\mathcal{R})$, these ‘stresses’ must further satisfy the equation

$$\frac{\Phi_1^{3/2}}{(1 + \Phi_1)^{1/2}} - \mathcal{R}^{-2} \frac{\Phi_0^{3/2}}{(1 + \Phi_0)^{1/2}} = \left[\sinh^{-1} \Phi^{1/2} - (1 + \Phi^{-1})^{-1/2} \right]_{\Phi_0}^{\Phi_1}. \quad (16d)$$

For a given indentation force \mathcal{F} , equations (16b) and (16d) form a pair of equations for the two unknowns Φ_0 and Φ_1 . Equation (16a) therefore yields an implicit force–displacement relation

$d(\mathcal{F}; \mathcal{R}, \nu)$. In the limit of the inner stress (or equivalently the radius) being taken to zero, $\Phi_0 = \mathcal{O}(\mathcal{R}^{4/3}) \rightarrow 0$, this system yields the point-indenter result of Vella and Davidovitch (2017, eq. (12)–(14)).

To make further analytical progress with a finite indenter radius, we concentrate on the two asymptotic limits $\mathcal{F} \rightarrow 0$ (§3.3.1) and $\mathcal{F} \rightarrow \infty$ (§3.3.2). This allows us to determine explicit forms that are formally valid only in these limits, but that might be expected to apply more broadly.

3.3.1. Moderate indentation forces ($\mathcal{F} \ll 1$)

The small indentation limit $\mathcal{F} \rightarrow 0$ is directly equivalent to taking $A[\Phi_0, \Phi_1] \rightarrow \infty$; consequently we find that $A \sim \Phi_1 = 4/\mathcal{F}^2 + \mathcal{O}(1)$ as $\mathcal{F} \rightarrow 0$. Inserting these into (16) gives the implicit force–displacement relation:

$$\frac{d}{\mathcal{F}} = \log \frac{4}{\mathcal{F}} - \sinh^{-1} \Phi_0^{1/2} + \mathcal{O}(\mathcal{F}^2), \quad (17a)$$

where Φ_0 is given by

$$\mathcal{R}^{-2} \frac{\Phi_0^{3/2}}{(1 + \Phi_0)^{1/2}} = \frac{4}{\mathcal{F}^2} + \mathcal{O}(1). \quad (17b)$$

The parameter Φ_0 can be eliminated from (17) to give an explicit equation for d in terms of \mathcal{F} ; however, the result is complicated and does not readily reveal the limiting behaviours. Instead we consider the sub-cases $\mathcal{F} \ll \mathcal{R}$ and $\mathcal{F} \gg \mathcal{R}$, which correspond to taking $\Phi_0 \rightarrow \infty$ and $\Phi_0 \rightarrow 0$, respectively.

At leading-order we find

$$\frac{d}{\mathcal{F}} \sim \begin{cases} \log \frac{1}{\mathcal{R}} & \text{for } \mathcal{F} \ll \mathcal{R} < 1, \\ \log \frac{4}{\mathcal{F}} & \text{for } \mathcal{R} \ll \mathcal{F} \ll 1. \end{cases} \quad (18)$$

The very small displacement solution in (18) (found when $\mathcal{F} \ll \mathcal{R}$) matches precisely with our earlier solution from (15) with $\mathcal{B} \ll \mathcal{R}^2$. Moreover, the moderate displacement solution in (18) (found when $\mathcal{R} \ll \mathcal{F} \ll 1$) is that presented by Vella and Davidovitch (2017) for a point indenter, and is shown as the red-dashed curve in Fig. 3. In summary, for a cylinder, as the indentation force increases, the indentation compliance d/\mathcal{F} evolves from being a constant (controlled by the indenter radius) to a logarithmic behaviour (controlled by an \mathcal{F} -dependent stretching-induced radius).

3.3.2. Large indentation forces ($\mathcal{F} \gg 1$)

The large indentation limit $\mathcal{F} \rightarrow \infty$ is equivalent to taking $A[\Phi_0, \Phi_1]/\Phi_1 \rightarrow 0$; consequently we find that $A = \mathcal{O}(\mathcal{F}^{-2/3})$ and $\Phi_1 = \mathcal{O}(1)$ as $\mathcal{F} \rightarrow \infty$. Inserting these into (16) gives the force–displacement relation:

$$\frac{d}{\mathcal{F}^{1/3}} = \frac{2}{(1 + \nu)^{1/3}} \frac{\sinh^{-1} \Phi_1^{1/2} - \sinh^{-1} \Phi_0^{1/2}}{\Phi_1^{1/2} (1 + \Phi_1)^{-1/6}} + \mathcal{O}(\mathcal{F}^{-2/3}), \quad (19a)$$

where Φ_1 and Φ_0 are the solutions to

$$\mathcal{R}^{-2} \frac{\Phi_0^{3/2}}{(1 + \Phi_0)^{1/2}} = \frac{(1 + \nu)}{2} \frac{\Phi_1^{3/2}}{(1 + \Phi_1)^{1/2}} + \mathcal{O}(\mathcal{F}^{-2/3}), \quad (19b)$$

$$\frac{1 - \nu}{2} \Phi_1 = \frac{(1 + \Phi_1)^{1/2}}{\Phi_1^{1/2}} \left[\sinh^{-1} \Phi_1^{1/2} - (1 + \Phi_1^{-1})^{-1/2} \right]_{\Phi_0}^{\Phi_1} + \mathcal{O}(\mathcal{F}^{-2/3}). \quad (19c)$$

The expression in (19a) corresponds to a constant cubic compliance: $d/\mathcal{F}^{1/3}$ is a function of the indenter size \mathcal{R} and Poisson's ratio ν alone.

Experimentally, the indenter is often orders of magnitude smaller than the sheet clamping radius, so that $\mathcal{R} \ll 1$ (Lee et al., 2008); this limit corresponds to $\Phi_0 = \mathcal{O}(\mathcal{R}^{4/3}) \rightarrow 0$, and so the cylindrical result (19) simplifies to

$$\frac{d}{\mathcal{F}^{1/3}} = \frac{2}{(1+\nu)^{1/3}} \frac{\sinh^{-1} \Phi_1^{1/2}}{\Phi_1^{1/2} (1+\Phi_1)^{-1/6}} - (2\mathcal{R})^{2/3} + \mathcal{O}(\mathcal{F}^{-2/3}, \mathcal{R}^2), \quad (20a)$$

where Φ_1 is the solution to

$$1 + \frac{1-\nu}{2} \Phi_1 = \frac{(1+\Phi_1)^{1/2}}{\Phi_1^{1/2}} \sinh^{-1} \Phi_1^{1/2} + \mathcal{O}(\mathcal{F}^{-2/3}, \mathcal{R}^2). \quad (20b)$$

This is the point-indenter result from Vella and Davidovitch (2017) with an additional $(2\mathcal{R})^{2/3}$ term to account for the small (but finite) indenter size. We find that the cubic compliance

$$\frac{d}{\mathcal{F}^{1/3}} \sim q_c[\nu, \mathcal{R}] = q_p[\nu] - (2\mathcal{R})^{2/3} + \mathcal{O}(\mathcal{R}^2), \quad (21)$$

where $q_c[\nu, \mathcal{R}]$ and $q_p[\nu]$ are given by the leading-order equations in (19) and (20) respectively. Thus, for large indentations by a cylindrical indenter ($\mathcal{B}^{1/2} \ll 1 \ll \mathcal{F}$), the cubic compliance $d/\mathcal{F}^{1/3}$ is a constant controlled by the sheet's Poisson's ratio ν and indenter radius \mathcal{R} . An analogous result was found by Vella and Davidovitch (2017, eq. (48)), for the case of a no-slip indenter: $d/\mathcal{F}^{1/3} \sim \alpha_0(\nu)^{-1/3} - [8\mathcal{R}^2/(1+\nu)]^{1/3}$ where $\alpha_0(\nu)^{-1/3} \equiv q_p[\nu]$. Figure 4 shows the comparison between numerical simulations, the prediction for a point indenter $\mathcal{R} = 0$, and the expression (21) for $\mathcal{R} = 0.1$. We see that the effect of finite indenter size is non-negligible in the large force limit.

The asymptotic predictions (15), (18), and (21) are presented in the regime diagram Fig. 2(a). We now move on from the case of a cylindrical indenter to one with a spherical tip.

4. Spherical indentation of a linearly elastic sheet

Having considered in some detail the simplest case of a finite cylindrical indenter, we now move on to a case of more experimental relevance: an indenter with a spherical tip (*e.g.* Bhatia and Nachbar 1968; Jennings et al. 1995; Begley and Mackin 2004; Lee et al. 2008; López-Polín et al. 2017). The key difference between this case and the cylindrical indenter already considered is that the radial position of the edge of contact, R_{in} , is initially unknown and evolves with the indentation depth, δ , as the sheet wraps more of the indenter.

4.1. Föppl-von Kármán formulation

The Föppl-von Kármán formulation for a spherical-capped indenter is the same as the cylindrical indenter formulation in §3.1 but with inner boundary conditions modified to account for the new indenter geometry. For consistency with the assumptions inherent in the FvK equations, we approximate the tip as a parabola. The analogue of the boundary conditions (12d–f) therefore take the dimensionless form

$$Z(\mathcal{R}) = -d + \frac{\mathcal{R}^2}{2\mathcal{R}_s}, \quad Z'(\mathcal{R}) = \frac{\mathcal{R}}{\mathcal{R}_s}, \quad Z''(\mathcal{R}) = \frac{1}{\mathcal{R}_s}, \quad (22a-c)$$

$$\Psi'(\mathcal{R}) - \frac{\Psi(\mathcal{R})}{\mathcal{R}} = -\frac{\mathcal{R}^2}{8\mathcal{R}_s^2}. \quad (22d)$$

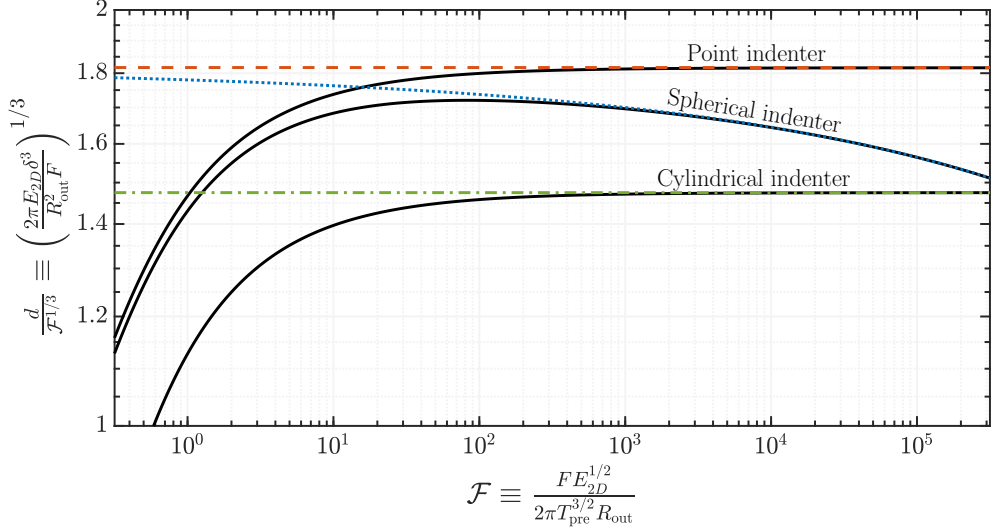


Figure 4: The cubic compliance associated with large indentations of a linear-elastic membrane ($\nu = 1/3$, $\mathcal{B} = 0$) for indenters of different type: results are shown for point ($\mathcal{R} \rightarrow 0$), cylindrical ($\mathcal{R} = 10^{-1}$), and spherical-capped ($\mathcal{R}_s = 10^{-3}$) indenters. Numerical solutions of the FvK equations are shown as solid black curves while the corresponding asymptotic solutions are shown as broken curves as follows: eq. (21)[$\mathcal{R} = 0$] in red (dashed), eq. (25a) in blue (dotted), and eq. (21) in green (dash-dotted). Observe the difference made by geometry: results for a spherical indenter never reach the constant cubic compliance regime $d/\mathcal{F}^{1/3} = \text{cst}$, while even for cylindrical indenters the finite indenter size may play a significant role.

Here (22a–c) express the geometric conditions of continuous vertical displacement, slope, and curvature, while (22d) comes from an in-plane force balance, since within the contact region it may easily be shown that $\Psi(r) + \rho^3/16\mathcal{R}_s^2 \propto \rho$. We again emphasize that the point of contact \mathcal{R} is unknown here and must be determined as part of the solution, explaining why we require an extra boundary condition (22c) compared to the cylindrical case.

The system of equations (11) subject to (12a–c) and (22) can be solved for given parameters ν , \mathcal{B} , \mathcal{F} , and \mathcal{R}_s by using a standard numerical integrator with unknown \mathcal{R} . To facilitate this computation, it is more convenient to fix the value of \mathcal{R} and solve for an unknown \mathcal{F} instead. By doing so, one avoids the problems associated with an unknown domain size.

To make analytical progress we apply the same asymptotic simplifications as in the cylindrical case: (i) linearize around the pre-stretched base state [*i.e.* take $\Psi(\rho) \sim \rho + \tilde{\Psi}(\rho)$ and $Z(\rho) \sim 0 + \tilde{Z}$ for $\tilde{\Psi}, \tilde{Z} \ll 1$]; or (ii) consider the membrane theory limit [*i.e.* take $\mathcal{B} \rightarrow 0$ and drop the boundary conditions (12b) and (22c)]. The analysis of these asymptotic limits are analogous to the cylindrical indenter case presented in §3.2 & §3.3, but with the added detail that \mathcal{R} is unknown; we shall only present the final results below (the full analysis can be found in Appendix A, for small indentation forces, and Appendix B, for small bending stiffnesses).

4.2. Small indentation forces ($\mathcal{F} \ll \max\{\mathcal{B}^{1/2}, \mathcal{R}_s\}$)

Linearizing around the pre-tensed state, we find that

$$\frac{d}{\mathcal{F}} \sim \begin{cases} \log \frac{e^\gamma}{2\mathcal{B}^{1/2}} & \text{for } \mathcal{R}_s \mathcal{F} \ll \mathcal{B} \ll 1, \\ \log \sqrt{\frac{e}{\mathcal{R}_s \mathcal{F}}} & \text{for } \mathcal{B} \ll \mathcal{R}_s \mathcal{F} < 1, \end{cases} \quad (23a)$$

with

$$\mathcal{R}^2 \sim \begin{cases} 4\mathcal{B}e^{-2\gamma} \exp\left[-\frac{4\mathcal{B}}{\mathcal{R}_s\mathcal{F}}\right] & \text{for } \mathcal{R}_s\mathcal{F} \ll \mathcal{B} \ll 1, \\ \mathcal{R}_s\mathcal{F} & \text{for } \mathcal{B} \ll \mathcal{R}_s\mathcal{F} < 1, \end{cases} \quad (23b)$$

where $\gamma \approx 0.577$ is again the Euler–Mascheroni constant. Thus, for small displacements by a spherical-capped indenter ($\mathcal{F} \ll \max\{\mathcal{B}^{1/2}, \mathcal{R}_s\}$), the indentation compliance d/\mathcal{F} evolves from being a constant (controlled by the bending-induced radius that was discussed in the cylindrical indenter problem) to a logarithmic behaviour (controlled by the radius of contact, which in turn depends on the force). This evolution is shown in Fig. 3 and confirms that the numerical results reproduce the expected asymptotic results in the relevant limits.

The asymptotic compliance (23a) is directly equivalent to that for a cylindrical indenter, *i.e.* (15), and can be recovered by accounting for the extra indentation depth due to the sphere geometry [$d \mapsto d + \mathcal{R}^2/2\mathcal{R}_s$, *i.e.* compare (12d) and (22a)] and inserting the contact radius expression (23b). It should be noted that the indenter-dominated solution (23)[$\mathcal{B} \ll \mathcal{R}_s\mathcal{F}$] has also been derived by Bhatia and Nachbar (1968, eq (43b)), from analytical solutions to small-rotation Reissner theory (analogous to FvK), and Norouzi et al. (2006, eq. (14)), by minimizing the energy of the constant-tension problem.

4.3. Moderate indentation forces ($\min\{\mathcal{B}^{1/2}, \mathcal{B}/\mathcal{R}_s\} \ll \mathcal{F} \ll 1$)

Taking the membrane theory limit and assuming small indentation forces ($\mathcal{F} \ll 1$) we find that

$$\frac{d}{\mathcal{F}} \sim \begin{cases} \log \sqrt{\frac{e}{\mathcal{R}_s\mathcal{F}}} & \text{for } \mathcal{F} \ll \mathcal{R}_s \ll 1, \\ \log \frac{4}{\mathcal{F}} & \text{for } \mathcal{R}_s \ll \mathcal{F} \ll 1, \end{cases} \quad (24a)$$

with

$$\mathcal{R}^2 \sim \begin{cases} \mathcal{R}_s\mathcal{F} & \text{for } \mathcal{F} \ll \mathcal{R}_s \ll 1, \\ 4(\sqrt{2} - 1)\sqrt{\mathcal{R}_s^3\mathcal{F}} & \text{for } \mathcal{R}_s \ll \mathcal{F} \ll 1. \end{cases} \quad (24b)$$

Thus, for moderate indentations by a spherical-capped indenter ($\min\{\mathcal{B}^{1/2}, \mathcal{B}/\mathcal{R}_s\} \ll \mathcal{F} \ll 1$), the indentation stiffness \mathcal{F}/d evolves from one logarithmic behaviour (controlled by the radius of contact) to another (controlled by a stretching-induced radius); these asymptotic results are also shown in Fig. 3.

Solution (24) is directly equivalent to the cylindrical result (18), and can again be obtained by accounting for the extra indentation depth due to the sphere geometry [$d \mapsto d + \mathcal{R}^2/2\mathcal{R}_s$, *i.e.* compare (12d) and (22a)] and inserting the contact radius expression (24b).

4.4. Large indentation forces ($\mathcal{F} \gg 1$)

Taking the membrane theory limit and assuming large indentation forces ($\mathcal{F} \gg 1$), we find that

$$\frac{d}{\mathcal{F}^{1/3}} \sim q_s[\nu, \mathcal{R}_s^3\mathcal{F}] = q_p[\nu] - 2(2 - \sqrt{2})(\mathcal{R}_s^3\mathcal{F})^{1/6} + \mathcal{O}(\sqrt{\mathcal{R}_s^3\mathcal{F}}), \quad (25a)$$

with

$$\mathcal{R}^2 \sim 4(\sqrt{2} - 1)\sqrt{\mathcal{R}_s^3\mathcal{F}}. \quad (25b)$$

Note that this result suggests a different effect of the geometry of the indenter, and the associated change in contact radius, than that proposed by Jia and Ben Amar (2020) based on a linear fitting procedure. We also emphasize that this result is different from the results presented by Begley and Mackin (2004) who neglect azimuthal strain and axial force balance to facilitate an approximate solution of the governing equations; in our notation this leads them to identify a constant cubic compliance, dependent on R_s/R_{out} . Instead, our asymptotic analysis shows that the cubic compliance is *not* constant but has a correction at $\mathcal{O}(\mathcal{R}_s^{1/2}\mathcal{F}^{1/6})$ that makes it weakly dependent on the force and indenter radius.

Above, we have implicitly assumed that $\mathcal{R}_s^3\mathcal{F} \ll 1$, which must be true for the assumption of small slopes to hold ($\mathcal{R} \ll \mathcal{R}_s \ll 1$). Thus, for large indentations by a spherical indenter ($\mathcal{F} \gg 1$), the cubic compliance $d/\mathcal{F}^{1/3}$ is controlled by the sheet’s Poisson’s ratio ν and contact radius $\mathcal{R} \sim (\mathcal{R}_s^3\mathcal{F})^{1/4}$. This is shown in Fig. 4, and demonstrates that in the spherical case the cubic compliance never saturates at a constant value (while it does for a cylindrical indenter). Nevertheless, the numerically-observed behaviour of the cubic compliance is in good agreement with the asymptotic prediction (25).

To our knowledge, the solution (25) is new. Moreover, while it is functionally similar to the corresponding cylindrical result (21), it cannot be obtained by accounting for the extra indentation depth [$d \mapsto d + \mathcal{R}^2/2\mathcal{R}_s$] and inserting the contact radius expression (25b), as was possible for small indentation depths. This difference is because the stress in the membrane that is in contact with the indenter is different between the two models — compare (12f) and (22d). The asymptotic results (23a), (24a), and (25a) are presented in the regime diagram Fig. 2(b).

4.5. Numerical results for a spherical indenter

| Reference | Material | $\frac{R_s}{R_{\text{out}}}$ | F_{max} [nN] | $\frac{R_{\text{out}}T_{\text{pre}}^{3/2}}{E_{2D}^{1/2}}$ [nN] | $E_{2D}R_s$ [nN] |
|---------------------------|------------------|------------------------------|-----------------------|--|------------------|
| Lee et al. (2008) | Graphene | 0.02 – 0.06 | 1200 – 2900 | 0.5 – 25.5 | 5600 – 9400 |
| Song et al. (2010) | h-BN | 0.05 | 200 | — | 1100 – 25500 |
| Bertolazzi et al. (2011) | MoS ₂ | 0.05 | 200 | 0.05 – 0.5 | 2200 |
| Lee et al. (2013) | Graphene | 0.03 – 0.08 | 2000 | 0.9 – 1.3 | 8600 – 12500 |
| López-Polín et al. (2017) | Graphene | 0.02 – 0.12 | 1200 – 2100 | 0.2 – 80 | 1500 – 4000 |
| Harats et al. (2020) | WS ₂ | 0.02 – 0.10 | 300 – 900 | 0.7 – 4.3 | 8500 |

Table 1: Typical experimental values of the parameters relevant to our model as determined in previous indentation experiments on various two-dimensional materials. (h-BN is hexagonal boron nitride, MoS₂ is molybdenum disulphide and WS₂ is tungsten disulphide.) Values of R_s/R_{out} are used to inform the parameters used in the numerical results shown in Fig. 5, while typical values of the maximum indentation force (denoted F_{max}) reached experimentally for graphene are indicated by the shaded region in Fig. 5. The values in the last two columns are pertinent to the discussion in §6.3, and are calculated using the reported values of E_{2D} and T_{pre} in the corresponding reference.

Figure 5 shows numerical results for the force–indentation relationship and the contact radius obtained from our FvK model with a spherical indenter. A common means of plotting experimental data is to plot the instantaneous estimate of $q(\nu)E_{2D}$ from the point indenter model, defined as $q(\nu)E_{2D} := FR_{\text{out}}^2/\delta^3$ with $q(\nu) := 2\pi q_p[\nu]^{-3}$ in our notation, as a function of applied force. Figure 5(a) mimics this by plotting $q(\nu)E_{2D}$ as a function of $F/(E_{2D}R_{\text{out}})$. Results are shown for a range of assumed pre-tensions and sphere radii, with the typical maximal applied loads used in previous AFM indentation experiments indicated (see Table 1 for details).

These numerical results show three particular features. Firstly, increasing the pre-tension may have the (undesired) effect of preventing measurements from approaching the horizontal asymptote before the maximum indentation force that can be applied is reached. Secondly, results with larger indenter radii of curvature, R_s/R_{out} , enhance the effect of the geometric nonlinearity, again pushing results further from the ideal point indenter solution of Schwerin (1929) and making it more difficult to infer the true value of E_{2D} . Thirdly, it seems that the fraction of the indenter that is wrapped by the membrane is relatively large, with R_{in}/R_s lying in the interval $0.3 \lesssim R_{\text{in}}/R_s \lesssim 0.7$ in the regime of experimental interest.

More importantly, however, the results of Fig. 5 show that the cubic compliance does not asymptote to a constant for spherical indenters with realistic material parameters, as assumed previously (see Figure 6 of Begley and Mackin, 2004, for example). Instead, the cubic compliance asymptotically decreases with increasing \mathcal{F} , as described by (25a) at large indentation forces. A decrease in cubic compliance corresponds to an increase in the instantaneous estimate of $E_{2D} \propto F/\delta^3$, as seen in Fig. 5(a); it therefore seems plausible that such non-constant behaviour might be interpreted experimentally as a nonlinear material effect, either softening or stiffening. Before discussing this possibility further, we turn now to consider such material nonlinearities.

5. Nonlinear elastic materials

Thus far, through the application of the Föppl-von Kármán equations, we have retained the leading-order geometrical nonlinearities associated with deformation, but have assumed that the material response remains Hookean: we have neglected any effect of nonlinear constitutive response. A simple energetic scaling (§2.3) shows that this assumption is valid provided that both $\mathcal{E} \ll 1$ and $\mathcal{F} \ll \mathcal{R}\mathcal{E}^{-3/2}$, where \mathcal{E} is the dimensionless pre-tension defined in (6). Many recent experiments have shown behaviour different to that expected on the basis of the FvK model of point indentation, and concluded that they are probing the nonlinear mechanical response of graphene (*e.g.* Lee et al. 2008; López-Polín et al. 2017). However, we have also seen that the geometry of a spherical indenter can give behaviour that differs from the usual cubic response (or constant cubic compliance) expected from the FvK equations. The question, therefore, is what happens when the indentation advances beyond the small-strain limit and how should one distinguish this regime from the geometrically nonlinear effects associated with indenter shape?

We introduce a model that allows for the possibility of large slopes and material strains. Specifically, we use the work of Green and Adkins (1960), who derived a generalized model for the large deformations of an elastic membrane. This formulation allows the membrane to have a stress-strain relationship that is nonlinear (and hence the solid is non-Hookean); we constrain the elastic constants introduced to recover the stretching modulus, E_{2D} , at small strains, and do not refer to an instantaneous effective stretching modulus at finite strains. The formulation presented below follows similar work by Yang and Feng (1970); Long et al. (2010); Pearce et al. (2011); Laprade et al. (2013) all of whom built upon Green and Adkins’s formulation.

We concentrate on the specific case of the indentation by a perfect-slip, spherical-capped wedge (with radius of curvature R_s and wedge angle $2\epsilon \ll 1$, as shown in Fig. 6). This type of indenter is commonly used in experiments (*e.g.* Lee et al. 2008; López-Polín et al. 2017) and reduces to the spherical-capped indenter used in §4 provided the indentation depth is sufficiently small. (Hence the results of this section should deviate from those of §4 only at large indentation depths.) Here, we also ignore the effects of the bending stiffness ($\mathcal{B} = 0$), since we are ultimately interested in the stretching dominant limit.

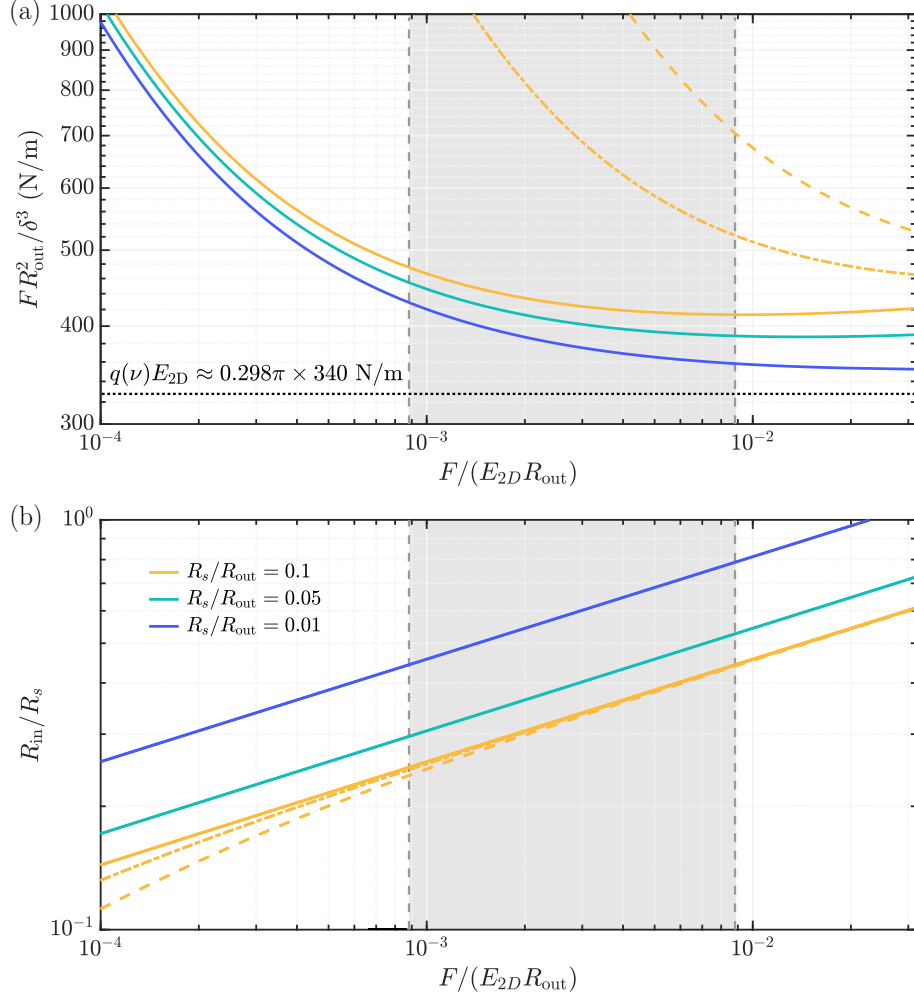


Figure 5: Numerical results obtained from the Föppl-von Kármán model with a spherical-capped indenter show the nonlinear effects associated with indenter geometry. (a) The instantaneous estimate of $q(\nu)E_{2D}(=FR_{out}^2/\delta^3)$ is plotted as a function of $F/(E_{2D}R_{out})$ for different values of R_s/R_{out} [indicated by colour: $R_s/R_{out} = 0.1$ (yellow), $R_s/R_{out} = 0.05$ (green) and $R_s/R_{out} = 0.01$ (blue)] and pre-tension T_{pre}/E_{2D} [indicated by line style: $T_{pre}/E_{2D} = 10^{-3}$ (solid curves), $T_{pre}/E_{2D} = 5 \times 10^{-3}$ (dash-dotted curves) and $T_{pre}/E_{2D} = 10^{-2}$ (dashed curves)]. The prediction of Schwerin (1929) for a point indenter with zero pre-tension is shown by the horizontal black dotted line. The typical range of experimental indentation forces used when fitting for the stretching modulus of graphene ($300 \text{ nN} \lesssim F \lesssim 3000 \text{ nN}$, as described in Table 1) is indicated by the shaded region under the assumption that $E_{2D}R_{out} = 3.4 \times 10^{-4} \text{ N}$. (b) The evolution of the radius at the edge of contact, R_{in}/R_s , is plotted as a function of force, with the same key as in (a) illustrating the varying pre-tension and sphere size. In both plots, true values $\nu = 0.165$ and $E_{2D} = 340 \text{ N/m}$ are assumed to present the numerical results in dimensional form.

5.1. Governing ODEs

To allow for large rotations of the sheet, it is useful to introduce intrinsic coordinates (the radial arc-length ξ and angle of rotation α , which is measured with respect to the radial- r -axis). We then have the geometrical conditions

$$\frac{dz}{d\xi} = \sin \alpha \quad \text{and} \quad \frac{dr}{d\xi} = \cos \alpha, \quad (26a,b)$$

where we recall that $0 \leq r \leq R_{\text{out}}$ and z are the radial and vertical coordinates of the sheet, respectively. These variables are sketched in Fig. 6.

The membrane is then split into two regions: the region in which the membrane contacts the tip and a non-contacting region. In the contacting region (which occupies $0 \leq r \leq R_{\text{in}}$ with R_{in} as yet unknown) we require

$$\sin \alpha = \begin{cases} \frac{r}{R_s} & \text{if } r \leq R_s \cos \epsilon, \\ \cos \epsilon & \text{if } r > R_s \cos \epsilon, \end{cases} \quad (26c)$$

for wedge angle 2ϵ . (Note that the wedge region is introduced to avoid the possibility that $r(\xi)$ becomes non-monotonic once the membrane inclination angle becomes close to $\pi/2$.) In the out-of-contact region (*i.e.* $R_{\text{in}} \leq r \leq R_{\text{out}}$) we impose the (integrated) vertical force balance

$$rT_\xi \sin \alpha = \frac{F}{2\pi}, \quad (26d)$$

where T_ξ and T_ϕ are the thickness-averaged, in-plane, radial and azimuthal stresses, which must satisfy the in-plane force balance

$$\frac{d}{dr} [rT_\xi] = T_\phi. \quad (26e)$$

Note that the out-of-plane stress is forced to be zero for thin sheets ($t \ll 1$) [Green and Adkins 1960].

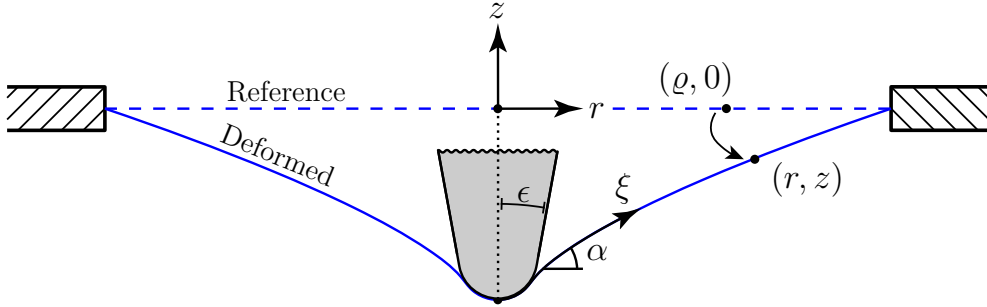


Figure 6: Cross-sectional sketch of the indentation of a clamped membrane showing the intrinsic and cylindrical coordinates (ξ, α) and (r, z) , respectively. The reference configuration of the membrane is illustrated by the dotted line and the deformed configuration by the solid curve. Note the different variables used to describe the reference and deformed configurations.

In modelling finite deformations of a thin membrane, one must distinguish between the reference and deformed configurations. Here, we take the reference configuration to be the planar sheet, subjected to an isotropic tension T_{pre} ; this configuration is parametrized by $(\varrho, 0)$ for $0 \leq \varrho \leq R_{\text{out}}$ (where clamping is imposed at $\varrho = r = R_{\text{out}}$). The variables describing the deformed configuration are expressed as functions of ϱ and hence we define the principal stretches as

$$\lambda_\xi := \frac{d\xi}{d\varrho}, \quad \lambda_\phi := \frac{r}{\varrho}, \quad \lambda_z := \frac{t}{t_0}. \quad (27a-c)$$

Here λ_ξ is the longitudinal stretch (along a cross-sectional curve in the r - z plane), λ_ϕ is latitudinal stretch (along the direction normal to r - z plane), and λ_z is the out-of-plane stretch (a measure of membrane thickness t compared to its reference value t_0). We also introduce the planar and

out-of-plane pre-stretches $\Lambda := \Lambda_\xi = \Lambda_\phi$ and Λ_z which measure the initial isotropic stretching of the sheet; we will relate these pre-stretches to the pre-tension T_{pre} shortly.

System (26) is closed by imposing a constitutive relation that links the stresses with the principal stretches (27). In particular, by assuming a hyperelastic isotropic medium, we can introduce a constitutive strain energy density (per unit-volume) function $W(\lambda_1, \lambda_2, \lambda_3)$ [see *e.g.* Holzapfel 2002] which directly links the stretches and stresses.

Using this formalism, we first calculate the pre-stretch Λ by solving the initial stress state,

$$\frac{\hat{t}_0}{\Lambda} \frac{\partial W}{\partial \lambda_1}(\Lambda, \Lambda, \Lambda_z) \equiv \frac{\hat{t}_0}{\Lambda} \frac{\partial W}{\partial \lambda_2}(\Lambda, \Lambda, \Lambda_z) = T_{\text{pre}}, \quad (28a)$$

with \hat{t}_0 being the undeformed sheet thickness and Λ_z chosen to satisfy the zero out-of-plane stress condition

$$\frac{\partial W}{\partial \lambda_3}(\Lambda, \Lambda, \Lambda_z) = 0. \quad (28b)$$

The stresses in the deformed configuration are then computed using

$$T_\xi = \frac{\hat{t}_0}{\Lambda \lambda_\phi} \frac{\partial W}{\partial \lambda_1}(\Lambda \lambda_\xi, \Lambda \lambda_\phi, \Lambda_z \lambda_z) \quad \text{and} \quad T_\phi = \frac{\hat{t}_0}{\Lambda \lambda_\xi} \frac{\partial W}{\partial \lambda_2}(\Lambda \lambda_\xi, \Lambda \lambda_\phi, \Lambda_z \lambda_z), \quad (29a,b)$$

where λ_z satisfies the zero out-of-plane stress condition

$$\frac{\partial W}{\partial \lambda_3}(\Lambda \lambda_\xi, \Lambda \lambda_\phi, \Lambda_z \lambda_z) = 0. \quad (29c)$$

Together, (26)–(29) form a system of three ordinary differential equations for the three unknowns — the in-plane stretches $\lambda_\xi(r)$ and $\lambda_\phi(r)$ and the vertical displacement $z(r)$:

$$\frac{d\lambda_\xi}{dr} = \frac{(T_\phi - T_\xi)\lambda_\xi \cos \alpha - \lambda_\phi T_{\xi, \phi}(\lambda_\xi \cos \alpha - \lambda_\phi)}{r \lambda_\xi T_{\xi, \xi} \cos \alpha}, \quad (30a)$$

$$\frac{d\lambda_\phi}{dr} = \left(1 - \frac{\lambda_\phi}{\lambda_\xi \cos \alpha}\right) \frac{\lambda_\phi}{r}, \quad (30b)$$

$$\frac{dz}{dr} = \tan \alpha, \quad (30c)$$

where $\alpha(r)$ is given by (26c) for $0 \leq r \leq R_{\text{in}}$ and (26d) for $R_{\text{in}} \leq r \leq R_{\text{out}}$; and $T_\xi(\lambda_\xi, \lambda_\phi)$, $T_\phi(\lambda_\xi, \lambda_\phi)$, and $T_{\xi, j}(\lambda_\xi, \lambda_\phi) := dT_\xi/d\lambda_j$ are given by (28) and (29). To proceed further requires a particular choice of strain energy function W , and so we turn to discuss this now.

5.2. Choice of strain energy density function

The choice of strain energy density function $W(\lambda_1, \lambda_2, \lambda_3)$ is informed by the material of interest. In this paper, we present results for two hyperelastic models to show the influence of this choice. In particular, we present results for a neo-Hookean material (the natural extension of the Hookean response that is implicit in the FvK equations) and for a Gent hyperelastic material (which is a model developed for polymeric materials with finite extensibility, but is chosen here as a qualitative way to account for the finite bond-lengths in graphene). The Gent model contains a parameter b that captures the finite chain length and recovers the neo-Hookean strain energy function as $b \rightarrow 0$ (corresponding to infinite chain extensibility). The formulation of the compressible strain energy

density functions is a lengthy process — the details are presented in Appendix C. The final form of the Gent energy density function used is

$$W = \frac{E}{4(1+\nu)} \left[-\frac{1}{b} \log [1 - b(I_1 - 3)] + (\beta - b)(I_3 - 1) - (1 + \beta - b) \log I_3 \right], \quad (31)$$

where $\beta := \nu/(1 - 2\nu)$ is some known constant, $\nu \neq 1/2$ is the Poisson ratio, I_i are the usual tensor invariants (defined in Appendix C), and b is an empirical parameter based on the finite extensibility of the material, defined such that $1/b \equiv \max\{I_1 - 3\}$.

5.3. Boundary conditions

Having split the domain into contacting ($0 \leq r \leq R_{\text{in}}$) and non-contacting regions ($R_{\text{in}} \leq r \leq R_{\text{out}}$), the problem is a multi-point boundary value problem and so we require boundary conditions at three positions $r = 0$, R_{in} , and R_{out} .

At the outer edge, $r = R_{\text{out}}$: the sheet is perfectly clamped,

$$\lambda_\phi(R_{\text{out}}) = 1 \quad \text{and} \quad z(R_{\text{out}}) = 0. \quad (32a,b)$$

At the origin, $r = 0$: we require an isotropic stretch (due to the symmetry of the problem),

$$\lambda_\phi(0) = \lambda_\xi(0) \quad \text{and} \quad z(0) = -\delta. \quad (32c,d)$$

At the interface between the contacting and non-contacting regions, $r = R_{\text{in}}$: a local force balance reveals that we require continuity in radial stress $T_\xi(\lambda_\xi, \lambda_\phi)$ and membrane slope α . Coupling this with the physical requirement of continuity of deformed variables z and $r = \varrho \lambda_\phi$, we require continuity in all our variables α , z , λ_ξ , and λ_ϕ . [This would not have been the case if there was a corner in the imposed indenter geometry (*e.g.* for a cylindrical punch).] Note that continuity in α gives an extra equation for the unknown point of contact $R_{\text{in}}(F)$.

5.4. Non-dimensionalization

To non-dimensionalize the problem we use the same choice of dimensionless radius and vertical coordinate (ρ and Z) as the Föppl-von Kármán formulation (10a,c), and define also

$$\rho := \frac{\varrho}{R_{\text{out}}}, \quad \hat{T}_\xi := \frac{T_\xi}{T_{\text{pre}}}, \quad \hat{T}_\phi := \frac{T_\phi}{T_{\text{pre}}}, \quad (33a-c)$$

and

$$\hat{W}(\lambda_\xi, \lambda_\phi, \lambda_z) := \frac{W(\lambda_\xi, \lambda_\phi, \lambda_z)}{E}, \quad (33d)$$

where we use $E_{2D} := \hat{t}_0 E$ to be the two-dimensional Young's Modulus of the undeformed sheet for small strains — this is equivalent to the FvK choice under small strains.

Substitution of (10a,c) and (33) into equations (26c,d), (30), and (32) gives a dimensionless system for $\lambda_\xi(\rho)$, $\lambda_\phi(\rho)$, $Z(\rho)$, and $\alpha(\rho)$, with Cauchy stresses given by

$$\hat{T}_\xi = \frac{\mathcal{E}^{-1}}{\Lambda \lambda_\phi} \frac{\partial \hat{W}}{\partial \lambda_1}(\Lambda \lambda_\xi, \Lambda \lambda_\phi, \Lambda_Z \lambda_z), \quad (34a)$$

$$\hat{T}_\phi = \frac{\mathcal{E}^{-1}}{\Lambda \lambda_\xi} \frac{\partial \hat{W}}{\partial \lambda_2}(\Lambda \lambda_\xi, \Lambda \lambda_\phi, \Lambda_Z \lambda_z), \quad (34b)$$

$$0 = \frac{\partial \hat{W}}{\partial \lambda_3}(\Lambda \lambda_\xi, \Lambda \lambda_\phi, \Lambda_Z \lambda_z), \quad (34c)$$

where the principal stretches Λ and Λ_z solve

$$\frac{1}{\Lambda} \frac{\partial \hat{W}}{\partial \lambda_1}(\Lambda, \Lambda, \Lambda_z) \equiv \frac{1}{\Lambda} \frac{\partial \hat{W}}{\partial \lambda_2}(\Lambda, \Lambda, \Lambda_z) = \mathcal{E} \quad \text{and} \quad \frac{\partial \hat{W}}{\partial \lambda_3}(\Lambda, \Lambda, \Lambda_z) = 0. \quad (34d,e)$$

For a given strain energy density function $\hat{W}(\lambda_1, \lambda_2, \lambda_3)$ [we use (31)] the above system can be solved by a standard numerical integrator (in our work we use `bvp4c` in `MATLAB`) for given parameters ν , \mathcal{F} , \mathcal{E} , and \mathcal{R}_s , with unknown \mathcal{R} . The associated indentation depth, $d(\mathcal{F})$, can then be calculated from (32d). In practice, however, it is more convenient to impose \mathcal{R} instead of \mathcal{F} and use the first derivative of (26d) to form a differential equation for the out-of-contact α :

$$\frac{d\alpha}{d\rho} = -\frac{T_\phi \tan \alpha}{\rho T_\xi}, \quad (35)$$

in $\mathcal{R} \leq \rho \leq 1$, with a continuity boundary condition at $\rho = \mathcal{R}$; \mathcal{F} can then be extracted post computation, along with d . By doing so, one avoids the issue of unknown domain size.

5.5. Results

The aim of introducing a nonlinear elastic model was to investigate when the effects of material (as opposed to geometric) nonlinearity are observed in the key force–displacement curve. Although the quantitative results are highly dependent on the chosen constitutive strain energy density function (31), the transition from the linearized-material asymptotics of §4 occurs at a similar indentation force, independent of the choice of strain energy function. We are therefore able to investigate numerically the effect of varying the sphere radius $\mathcal{R}_s = (R_s/R_{\text{out}})\mathcal{E}^{1/2}$ and pre-tension \mathcal{E} . In Fig. 7 we present a table of force–displacement curves obtained as \mathcal{E} and \mathcal{R}_s vary; observe that the dimensionless force at which the results deviate from the Föppl-von Kármán solutions increases with R_s/R_{out} and decreases with $\mathcal{E} = T_{\text{pre}}/E_{2D}$ — reminiscent of the prediction from the energetic analyses, $\mathcal{F} \sim \mathcal{R}_s/\mathcal{E}^2$, presented in §2.2. It is also interesting to note that, close to the place at which the non-Hookean results deviate from the FvK results, the effect of material nonlinearity is to soften the material response. This is in contrast to the geometrical nonlinearities discussed in §4, which acted to increase the instantaneous estimate of E_{2D} , *i.e.* to stiffen the indentation response. We discuss these results, and their significance for indentation probes of the elastic constants of thin materials, now.

6. Discussion: Application to fitting protocols

The controlled indentation of thin sheets is a common, but delicate, experimental technique used to extract mechanical properties of thin, approximately two-dimensional, materials. Our detailed analysis of the cylindrical and spherical indentation has led to a number of asymptotic results that highlight the complexity of this problem (these results are summarized in §7). Consequently, there are a number of potential pitfalls in this fitting procedure that must be appreciated if they are to be avoided and a reliable measurement of the quantity of interest to be obtained. Below, we discuss these pitfalls in the context of measurements of three properties: the sheet pre-tension T_{pre} , the two-dimensional Young’s modulus E_{2D} , and the non-Hookean material behaviour (*i.e.* the behaviour not governed by linear elasticity). We shall concentrate on spherical-capped indenters, since these are among the most commonly used in practice.

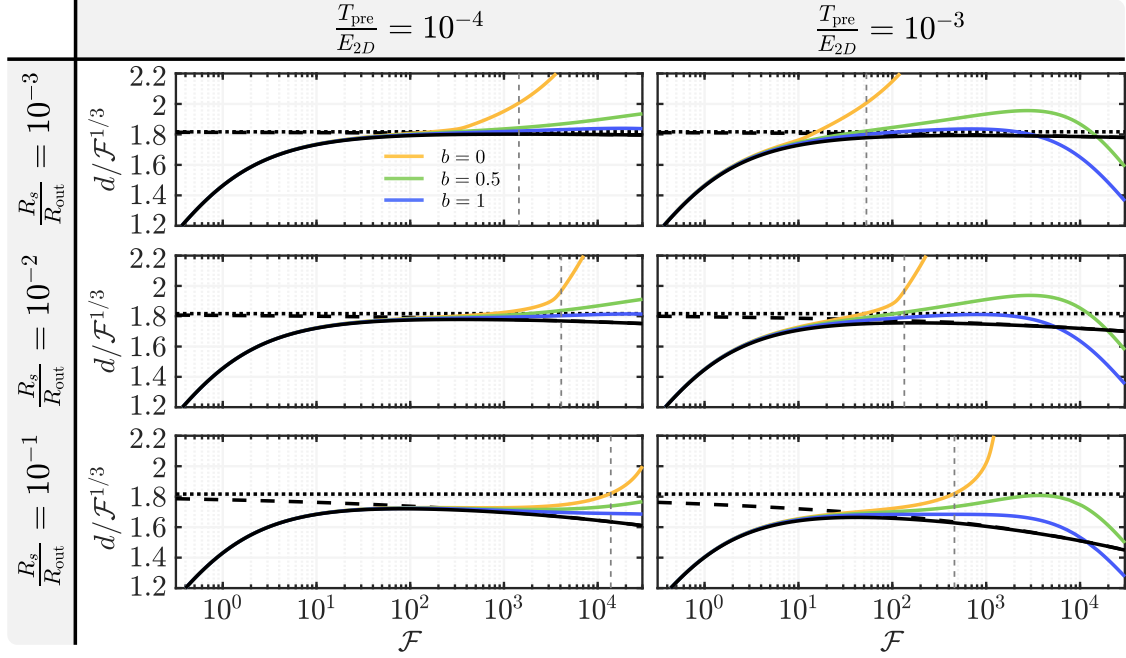


Figure 7: Table of plots showing how the onset of nonlinear elasticity depends on the spherical cap curvature (rows) and the applied pre-tension (columns) of the membrane ($\nu = 1/3$, $\mathcal{B} = 0$) — here we have used an indenter wedge angle of $2\epsilon = \pi/9$ (matching the value in *e.g.* López-Polín et al. 2017). Numerical solutions are shown as solid curves: Föppl-von Kármán (linear elastic) in black, Gent ($b = 1$) in blue, Gent ($b = 0.5$) in green, and neo-Hookean ($b = 0$) in yellow. The asymptotic results from the FvK analysis for a point and spherical indenter, (25a) with $\mathcal{R}_s = 0$ and $\mathcal{R}_s = (R_s/R_{\text{out}})(T_{\text{pre}}/E_{2D})^{1/2}$, are shown as dotted and dashed lines respectively. The vertical dashed lines show where the relative difference between the Neo-Hookean and FvK models reaches 10%.

6.1. Measuring sheet pre-tension

Our results showed the existence of a small-indentation regime in which the pre-tension dominates and $F \propto T_{\text{pre}}\delta$ (Fig. 2). Based on our analysis of this region, there are two potential pitfalls that may cause errors when attempting to infer T_{pre} from experimental measurements of F :

- *Bending stiffness.* Although the bending stiffness is small at a macroscopic level, *i.e.* $B \ll T_{\text{pre}}R_{\text{out}}^2$, it is not necessarily negligible in the early stages of indentation with a spherical-capped indenter (see Fig. 2(b)): at small indentations, the spherical cap is barely wrapped by the sheet, the contact radius is small and it is the bending stiffness of the sheet that dominates through an effective, bending-induced, radius $R_{\text{in}}^{\text{eff}} \sim (B/T_{\text{pre}})^{1/2}$, instead of the contact radius. Avoiding this requires that the applied force be sufficiently large; in particular, that $F \gg B/R_s$ for small bending stiffnesses ($B^{1/2} \ll R_s T_{\text{pre}}/E_{2D}^{1/2}$) and $F \gg B^{1/2} T_{\text{pre}}/E_{2D}^{1/2}$ for moderate bending stiffnesses ($R_s T_{\text{pre}}/E_{2D}^{1/2} \ll B^{1/2} \ll R_{\text{out}} T_{\text{pre}}^{1/2}$).
- *Sheet stretching.* If the indentation depths used are not sufficiently small, the pre-tension might be insignificant compared to the tension associated with indentation-induced stretching. This would cause a non-linear response (*i.e.* the sheet transitions from the linear $\mathcal{F} \sim d$ towards the cubic $\mathcal{F} \sim d^3$ behaviour). Avoiding this requires that the indentation force is not too large, in particular that $F \ll R_{\text{out}} T_{\text{pre}}^{3/2}/E_{2D}^{1/2}$.

Combining these requirements, we find that indentation tests aimed at measuring the pre-tension in a sheet should focus on indentation forces $F_{T_{\text{pre}}}$ such that

$$\min \left\{ \frac{B}{R_s}, \frac{B^{1/2} T_{\text{pre}}}{E_{2D}^{1/2}} \right\} \ll F_{T_{\text{pre}}} \ll \frac{R_{\text{out}} T_{\text{pre}}^{3/2}}{E_{2D}^{1/2}}. \quad (36)$$

6.2. Measuring 2D Young's modulus

The measurement of the two-dimensional Young's modulus of a sheet requires experiments to be performed in the stretching dominated regime, where $F \propto E_{2D} \delta^3 / R_{\text{out}}^2$ (Fig. 2). Based on our analysis of this regime, there are two immediate potential pitfalls that may cause fitting errors:

- *Sheet pre-tension.* If the sheet is not sufficiently indented, the effect of the pre-tension may still be significant, leading to a non-cubic response (*i.e.* the sheet is still transitioning between the linear $\mathcal{F} \sim d$ and cubic $\mathcal{F} \sim d^3$ behaviours, see Fig. 7). To avoid this, sufficiently large forces should be applied; in particular $F \gg R_{\text{out}} T_{\text{pre}}^{3/2} / E_{2D}^{1/2}$.
- *Mechanical nonlinearities.* If the sheet is indented too much, the stress in the sheet exceeds that for which the linear elastic (Hookean) constitutive response is valid, and nonlinearities become important. In this case, the expected cubic response may not be observed (see the solution divergence in Fig. 7) causing errors in the fitted value of E_{2D} . To ensure this possibility is avoided requires $F \ll E_{2D} R_s$.

Combining these requirements, we find that indentation tests aimed at measuring the stretching modulus of a sheet should focus on indentation forces $F_{E_{2D}}$ such that

$$\frac{R_{\text{out}} T_{\text{pre}}^{3/2}}{E_{2D}^{1/2}} \ll F_{E_{2D}} \ll E_{2D} R_s. \quad (37)$$

A third potential pitfall is less obvious and independent of the indentation force applied:

- *Geometrical nonlinearities.* When fitting experimental data to obtain a value for the 2D Young's modulus, it is common practice to use the Schwerin (1929) point indenter solution, which in our notation reads $\mathcal{F} = Q(\nu) d^3$ for $Q(\nu) := q_p[\nu]^{-3}$. For example, a point-wise estimate $E_{2D} = F R_{\text{out}}^2 / [2\pi Q(\nu) \delta^3]$ is often used (*e.g.* in López-Polín et al. 2017). However, the analysis presented here shows that the indenter geometry is important and may lead to large errors in the fitted value of the 2D Young's modulus if not accounted for. In particular, for spherical indenters the point-wise estimate of E_{2D} may not converge as the force increases — the constant value (as predicted by Schwerin) is never observed (see Figs 4 & 7). To avoid a significant effect of the spherical geometry requires $R_s / R_{\text{out}} \ll 1$; however, we caution that decreasing R_s or increasing R_{out} may have the undesired effect of decreasing the desired range of indentation forces in (37), risking other fitting errors. In practice, $R_s / R_{\text{out}} \lesssim 10^{-2}$ appears to be sufficient (see Fig. 7); in many experimental setups $R_s / R_{\text{out}} \sim 0.1$ (see López-Polín et al., 2017, for example) suggesting that indenter geometry may play a role in interpreting previous experimental results.

6.3. Measuring non-Hookean material behaviour

If interested in examining the non-Hookean material behaviour of the sheet (*i.e.* the behaviour beyond the Hookean linear stress-strain relation), there are two potential pitfalls to avoid:

- *Remaining in the linear elastic regime.* The main concern when measuring non-Hookean behaviour is whether the strains induced by indentation are large enough to be controlled by a nonlinear constitutive law. For a spherical-capped indenter, this requires

$$E_{2D}R_s \ll F. \quad (38)$$

- *Geometrical nonlinearities.* A less obvious trap is that deviating from Schwerin's solution [$\mathcal{F} = Q(\nu)d^3$] might be interpreted as nonlinear mechanical effects, but actually result from nonlinear geometry. For example, for sufficiently large spherical indenters, even the Föppl-von Kármán response will never achieve the plateau expected from the Schwerin result (see Figs 4 & 7). This failure to reach the Schwerin regime might be interpreted as a material nonlinearity, rather than a universal geometric property that is predicted by Hookean elasticity. To avoid this possibility requires geometric effects to be negligible throughout the linear elastic regime, that is $R_s \ll R_{\text{out}}$. However, as we now discuss, our results suggest that the effect of geometrical and material nonlinearities on the cubic compliance $d/\mathcal{F}^{1/3}$ are qualitatively different.

6.4. Distinguishing geometrical and material nonlinearities

Together, (37) and (38) provide limits on the validity of fitting force-indentation curves to determine the stretching modulus E_{2D} of a two-dimensional material, and to distinguish between geometrical and material nonlinearities in this procedure. (Of course, their validity can only be checked with some initial parameter estimate or *a posteriori*.) It should be noted that these are only based on order arguments and are not concrete cut off points. For example, the numerical results presented in Figs 4 & 7 suggest that to observe the asymptotic results for $\mathcal{F} \gg 1$ in practice requires $\mathcal{F} \gtrsim 100$; hence a useful guide for satisfying the conditions (37) in practice is

$$200\pi \times \frac{R_{\text{out}}T_{\text{pre}}^{3/2}}{E_{2D}^{1/2}} \lesssim F_{E_{2D}} \ll E_{2D}R_s. \quad (39)$$

Comparison of the relevant experimental parameters collected in Table 1 with (39) suggests that the maximum indentation force applied experimentally, F_{max} , does not always reach the large multiple of $T_{\text{pre}}^{3/2}R_{\text{out}}/E_{2D}^{1/2}$ required for accurate measures of E_{2D} from asymptotic results.

Hence, when fitting, one should always ensure the desired asymptotic behaviour is observed. To fit the stretching stiffness E_{2D} , this might be most simply done by ensuring that there is indeed a cubic plateau, $F/d^3 \sim \text{constant}$. However, our results show that a true plateau is only obtained with a cylindrical indenter, while most experiments use a shape that is closer to a spherical cap. Fortunately, qualitatively similar asymptotic results may be derived from the Föppl-von Kármán equations for a spherical-capped indenter; in this regard (25a) might be expected to be especially helpful and so we note that it may be rewritten in dimensional terms as

$$\frac{\delta}{R_{\text{out}}} \sim \frac{q_p[\nu]}{(2\pi)^{1/3}} \left(\frac{F}{E_{2D}R_{\text{out}}} \right)^{1/3} - \frac{2(\sqrt{2}-1)}{\pi^{1/2}} \left(\frac{R_s}{R_{\text{out}}} \right)^{1/2} \left(\frac{F}{E_{2D}R_{\text{out}}} \right)^{1/2}. \quad (40)$$

Note that (40) reduces to the classical Schwerin result as $R_s/R_{\text{out}} \rightarrow 0$ but shows that the perturbation caused by spherical geometry is *not* of the prefactor (as assumed by Begley and Mackin, 2004, for example) but rather is additive. Moreover, (40) shows that the cubic compliance $\delta/F^{1/3}$ is decreased by the effect of the indenter's radius of curvature R_s : the geometry of the indenter means that the apparent stiffness of the suspended solid is *increased* compared to a point indenter, and increases further with increasing load. As a rule of thumb, our results suggest that material nonlinearities tend to soften the response initially (at least for the strain energy functionals considered here) while geometrical nonlinearities tend to stiffen the response.

7. Summary of results and conclusions

7.1. Summary

We have presented a series of asymptotic solutions that may be used in combination with experimental force-indentation data to fit mechanical properties of thin materials, subject to an appreciation of the pitfalls described in §6. These results apply in various asymptotic regimes determined by the relative sizes of the dimensionless force \mathcal{F} , bending stiffness \mathcal{B} , indenter radius \mathcal{R} (or \mathcal{R}_s), and pre-tension \mathcal{E} . Here, we summarize these results for a cylindrical and spherical-capped indenter; these are most easily expressed in terms of the vertical indentation depth d achieved for a fixed force \mathcal{F} .

Cylindrical indenter. For indentation by a cylindrical indenter, the asymptotic response is dependent on the relative size of the bending stiffness \mathcal{B} and indenter area \mathcal{R}^2 . We have that

$$\text{if } \mathcal{B}^{1/2} \ll \mathcal{R} < 1: \quad d \sim \begin{cases} \mathcal{F} \log \frac{1}{\mathcal{R}} & \text{for } 0 \leq \mathcal{F} \ll \mathcal{R}, \\ \mathcal{F} \log \frac{4}{\mathcal{F}} & \text{for } \mathcal{R} \ll \mathcal{F} \ll 1, \\ \mathcal{F}^{1/3} q_c[\nu, \mathcal{R}] & \text{for } 1 \ll \mathcal{F}, \end{cases} \quad (41a)$$

whilst

$$\text{if } \mathcal{R} \ll \mathcal{B}^{1/2} \ll 1: \quad d \sim \begin{cases} \mathcal{F} \log \frac{e^\gamma}{2\mathcal{B}^{1/2}} & \text{for } 0 \leq \mathcal{F} \ll \mathcal{B}^{1/2}, \\ \mathcal{F} \log \frac{4}{\mathcal{F}} & \text{for } \mathcal{B}^{1/2} \ll \mathcal{F} \ll 1, \\ \mathcal{F}^{1/3} q_c[\nu, \mathcal{R}] & \text{for } 1 \ll \mathcal{F}. \end{cases} \quad (41b)$$

Here, $\gamma \approx 0.577$ is the Euler–Mascheroni constant (Abramowitz and Stegun, 1964) and $q_c[\nu, \mathcal{R}] \sim q_p[\nu] - (2\mathcal{R})^{2/3}$ is defined in (21). These solutions are valid provided $\mathcal{E} \ll 1$ and $\mathcal{F} \ll \mathcal{R}\mathcal{E}^{-3/2}$; if either of these conditions fail then the material instead behaves according to a nonlinear constitutive law (non-Hookean behaviour). The regions of the regime diagram Fig. 2(a) are delineated by the expressions in (41).

Spherical indenter. For indentation by a spherical-capped indenter, we find the asymptotic response:

$$d \sim \begin{cases} \mathcal{F} \log \frac{e^\gamma}{2\mathcal{B}^{1/2}} & \text{for } 0 \leq \mathcal{F} \ll \min\{\mathcal{B}/\mathcal{R}_s, \mathcal{B}^{1/2}\}, \\ \mathcal{F} \log \sqrt{\frac{e}{\mathcal{R}_s \mathcal{F}}} & \text{for } \mathcal{B}/\mathcal{R}_s \ll \mathcal{F} \ll \mathcal{R}_s, \\ \mathcal{F} \log \frac{4}{\mathcal{F}} & \text{for } \max\{\mathcal{R}_s, \mathcal{B}^{1/2}\} \ll \mathcal{F} \ll 1, \\ \mathcal{F}^{1/3} q_s[\nu, \mathcal{R}_s^3 \mathcal{F}] & \text{for } 1 \ll \mathcal{F}, \end{cases} \quad (42a)$$

with the (*a priori* unknown) contact radius given by

$$\mathcal{R}^2 \sim \begin{cases} 4\mathcal{B}e^{-2\gamma} \exp[-4\mathcal{B}/\mathcal{R}_s \mathcal{F}] & \text{for } 0 \leq \mathcal{F} \ll \min\{\mathcal{B}/\mathcal{R}_s, \mathcal{B}^{1/2}\}, \\ \mathcal{R}_s \mathcal{F} & \text{for } \mathcal{B}/\mathcal{R}_s \ll \mathcal{F} \ll \mathcal{R}_s, \\ 4(\sqrt{2} - 1)\mathcal{R}_s^{3/2} \mathcal{F}^{1/2} & \text{for } \max\{\mathcal{R}_s, \mathcal{B}^{1/2}\} \ll \mathcal{F}. \end{cases} \quad (42b)$$

Here, $q_s[\nu, \mathcal{R}_s^3 \mathcal{F}] \sim q_p[\nu] - 2(2 - \sqrt{2})(\mathcal{R}_s^3 \mathcal{F})^{1/6}$ is defined in (25) and we note that the dimensional version of (42a) for $\mathcal{F} \gg 1$ is given in (40). These solutions are valid provided $\mathcal{E} \ll 1$ and $\mathcal{F} \ll \mathcal{R}_s \mathcal{E}^{-2}$; if either of these conditions fail then the material instead behaves according to a nonlinear constitutive law (non-Hookean behaviour). The regions of the regime diagram Fig. 2(b) are delineated by the expressions in (42).

7.2. Conclusions

Altogether, our work provides a comprehensive description of the Hookean response of a sheet subject to localized indentation accounting for geometrical nonlinearities, while additionally providing information about when a non-Hookean response can be expected. It is common in experiments to use dimensional versions of similar asymptotic solutions to extract information about the material of interest (*e.g.* its 2D Young's modulus, E_{2D} , or pre-tension, T_{pre}). The asymptotic results presented in (41) and (42) show the number of different regimes that exist and hence the difficulty of choosing the appropriate asymptotic result. Nevertheless, understanding the appropriate regime for each of these results is important, since incorrect choices may lead to large errors in the fitted values obtained. For instance, if the indenter was assumed to be point-like (so that $q_c[\nu, \mathcal{R}] \approx q_p[\nu]$ or $q_s[\nu, \mathcal{R}_s^3 \mathcal{F}] \approx q_p[\nu]$), we would obtain $\mathcal{O}(\mathcal{R}^{2/3})$ or $\mathcal{O}(\mathcal{R}_s^{1/2} \mathcal{F}_{\text{max}}^{1/6})$ errors in the fitted Young's modulus — which can be significant in such a sensitive process. We therefore emphasize the importance of using the correct response when fitting parameters and suggest the use of (40) to account for indenters with a hemi-spherical tip.

Finally, we note that the asymptotic regimes considered here have been motivated by recent indentation experiments on ultra-thin materials, including few layer graphene. For such materials, $B \ll T_{\text{pre}} R_{\text{out}}^2$, corresponding to $\mathcal{B} \ll 1$, and so the different regimes described in eqns (41) and (42) are only valid when $\mathcal{B} \ll 1$. As the dimensionless bending stiffness becomes larger, $\mathcal{B} = \mathcal{O}(1)$, the balances that lead to these results are expected to change. In particular, we expect that for dimensionless forces $\mathcal{F} \lesssim 1$ and $\mathcal{B} \sim 1$ new results would be required (indicated by the greyed-out regions in Fig. 2).

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Appendix A. Asymptotic solutions for small indentation forces

In this Appendix, we extend the arguments of §3.2 and §4.2 to derive the asymptotic solutions (15) and (23). We shall present the analysis for the cylindrical and spherical-capped indenters simultaneously.

We consider a small perturbation of the initial pre-tensed configuration: $\Psi(\rho) \sim \rho + \tilde{\Psi}(\rho)$ and $Z(\rho) \sim \tilde{Z}(\rho)$, anticipating that $\tilde{\Psi}, \tilde{Z} \ll 1$. At leading order (in $\tilde{\Psi}$ and \tilde{Z}), the Föppl-von Kármán equations (11) take the form,

$$\mathcal{B}\rho \frac{d}{d\rho} \left[\frac{1}{\rho} \frac{d}{d\rho} \left(\rho \frac{d\tilde{Z}}{d\rho} \right) \right] = \rho \frac{d\tilde{Z}}{d\rho} - \mathcal{F}, \quad (\text{A.1a})$$

$$\rho \frac{d}{d\rho} \left[\frac{1}{\rho} \frac{d}{d\rho} (\rho \tilde{\Psi}) \right] = -\frac{1}{2} \left(\frac{d\tilde{Z}}{d\rho} \right)^2. \quad (\text{A.1b})$$

Most importantly (A.1a) no longer couples the out-of-plane deflection with the in-plane stress; the third order differential equation for the sheet profile $Z(\rho) \sim \tilde{Z}(\rho)$ can immediately be integrated to give

$$Z \sim \tilde{Z} = c_1 I_0 \left[\frac{\rho}{\mathcal{B}^{1/2}} \right] + c_2 K_0 \left[\frac{\rho}{\mathcal{B}^{1/2}} \right] + \mathcal{F} \log \rho + c_3, \quad (\text{A.2})$$

with the constants c_1 , c_2 , and c_3 yet to be determined. (The stress profile may be determined by substituting (A.2) into (A.1b); we omit this here since it affects the force-indentation response only at higher order in d .)

The sheet profile (A.2) is to be solved subject to the boundary conditions

$$Z(1) = 0, \quad Z'(1) = 0, \quad (\text{A.3a,b})$$

with

$$Z(\mathcal{R}) = -d, \quad Z'(\mathcal{R}) = 0, \quad (\text{A.4a,b})$$

for a cylindrical indenter (\mathcal{R} known), or

$$Z(\mathcal{R}) = -d + \frac{\mathcal{R}^2}{2\mathcal{R}_s}, \quad Z'(\mathcal{R}) = \frac{\mathcal{R}}{\mathcal{R}_s}, \quad Z''(\mathcal{R}) = \frac{1}{\mathcal{R}_s}, \quad (\text{A.5a-c})$$

for a spherical indenter (\mathcal{R} unknown). Implementation of (A.3)–(A.5) gives explicit expressions for the coefficients

$$\frac{c_1}{\mathcal{F}\mathcal{B}^{1/2}} = \frac{\hat{K}_1^R - \hat{K}_1/\mathcal{R}}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R}, \quad \frac{c_2}{\mathcal{F}\mathcal{B}^{1/2}} = \frac{\hat{I}_1^R - \hat{I}_1/\mathcal{R}}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R}, \quad (\text{A.6a,b})$$

and

$$c_3 = -c_1 \hat{I}_0 - c_2 \hat{K}_0, \quad (\text{A.6c})$$

for a cylindrical indenter. For a spherical indenter, we have

$$\frac{c_1}{\mathcal{F}\mathcal{B}^{1/2}} = \frac{\hat{K}_1 \mathcal{R} / \mathcal{F}\mathcal{R}_s + \hat{K}_1^R - \hat{K}_1 / \mathcal{R}}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R}, \quad \frac{c_2}{\mathcal{F}\mathcal{B}^{1/2}} = \frac{\hat{I}_1 \mathcal{R} / \mathcal{F}\mathcal{R}_s + \hat{I}_1^R - \hat{I}_1 / \mathcal{R}}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R}, \quad (\text{A.7a,b})$$

and

$$c_3 = -c_1 \hat{I}_0 - c_2 \hat{K}_0, \quad (\text{A.7c})$$

with \mathcal{R} implicitly given by

$$\frac{1}{\mathcal{R}_s} + \frac{\mathcal{F}}{\mathcal{R}^2} = \frac{1}{2\mathcal{B}} \left[c_1 \left(\hat{I}_0^R + \hat{I}_2^R \right) + c_2 \left(\hat{K}_0^R + \hat{K}_2^R \right) \right]. \quad (\text{A.7d})$$

(In each case, \hat{I}_j , \hat{I}_j^R , \hat{K}_j , and \hat{K}_j^R are as defined in eq. (14).)

The small force-displacement relations are then found by imposing the boundary conditions (A.4a) and (A.5a), to give:

$$\frac{d}{\mathcal{F}} = \log \frac{1}{\mathcal{R}} + \frac{\hat{K}_1 \hat{I}_0^R + \hat{I}_1 \hat{K}_0^R + \mathcal{R} \left(\hat{K}_0 \hat{I}_1^R + \hat{I}_0 \hat{K}_1^R \right) - 2\mathcal{B}^{1/2} \mathcal{B}^{1/2}}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R} \frac{\mathcal{B}^{1/2}}{\mathcal{R}}, \quad (\text{A.8})$$

for a cylindrical indenter; and

$$\begin{aligned} \frac{d}{\mathcal{F}} = \log \frac{1}{\mathcal{R}} + & \frac{\hat{K}_1 \hat{I}_0^R + \hat{I}_1 \hat{K}_0^R + \mathcal{R} \left(\hat{K}_0 \hat{I}_1^R + \hat{I}_0 \hat{K}_1^R \right) - 2\mathcal{B}^{1/2} \mathcal{B}^{1/2}}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R} \frac{\mathcal{B}^{1/2}}{\mathcal{R}} \\ & + \frac{\mathcal{B}^{1/2} - \hat{I}_1 \hat{K}_0^R - \hat{I}_0 \hat{K}_1}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R} \frac{\mathcal{R} \mathcal{B}^{1/2}}{\mathcal{R}_s \mathcal{F}} + \frac{\mathcal{R}^2}{2\mathcal{R}_s \mathcal{F}}, \end{aligned} \quad (\text{A.9a})$$

with \mathcal{R} implicitly given by

$$\frac{\mathcal{F}\mathcal{R}_s + \mathcal{B}^{-1/2} (\mathcal{R}^2 - \mathcal{F}\mathcal{R}_s) \left(\hat{I}_0^R \hat{K}_1 + \hat{K}_0^R \hat{I}_1 \right)}{\hat{I}_1^R \hat{K}_1 - \hat{I}_1 \hat{K}_1^R} = 2\mathcal{R}, \quad (\text{A.9b})$$

for a spherical indenter.

In the limit of small bending stiffnesses which is relevant here, $\mathcal{B} \ll 1$, (A.8) and (A.9) can be simplified using the asymptotic behaviour of modified Bessel functions to give

$$\frac{d}{\mathcal{F}} = \log \frac{1}{\mathcal{R}} - \mathcal{B}^{1/2} \frac{\hat{K}_0^R}{\mathcal{R} \hat{K}_1^R} - \mathcal{B}^{1/2} \frac{\hat{I}_0}{\hat{I}_1} + \text{e.s.t.}, \quad (\text{A.10})$$

for a cylindrical indenter. For a spherical indenter

$$\frac{d}{\mathcal{F}} = \log \frac{1}{\mathcal{R}} - \frac{2\mathcal{B}}{\mathcal{R}_s \mathcal{F}} + \frac{\mathcal{R}^2}{2\mathcal{R}_s \mathcal{F}} - \mathcal{B}^{1/2} \frac{\hat{I}_0}{\hat{I}_1} + \text{e.s.t.}, \quad (\text{A.11a})$$

where \mathcal{R} is given implicitly by

$$(\mathcal{R}_s \mathcal{F} - \mathcal{R}^2) \frac{\hat{K}_0^R}{\mathcal{R} \hat{K}_1^R} = 2\mathcal{B}^{1/2} + \text{e.s.t.}. \quad (\text{A.11b})$$

Finally, considering the limits $\mathcal{B} \ll \mathcal{R}^2 < 1$ and $\mathcal{R}^2 \ll \mathcal{B} \ll 1$ leads to the leading-order solutions (15) for a cylindrical indenter and (23) for a spherical-capped indenter.

Appendix B. Asymptotic solutions for small bending stiffnesses

In this Appendix, we expand the arguments of §3.3, §4.3, and §4.4 to derive the asymptotic solutions for large indentation depths, and negligible bending stiffnesses (18), (21), (24), and (25). We follow the analysis of a point-indenter by Vella and Davidovitch (2017, App. B) and present the case of cylindrical and spherical-capped indenters simultaneously.

Following the discussion in §3.3, we let $\eta = \rho^2$, $\Phi = \rho\Psi$ and set $\mathcal{B} = 0$ in (11), to give:

$$2\Phi \frac{dZ}{d\eta} = \mathcal{F}, \quad (\text{B.1a})$$

$$\frac{d^2\Phi}{d\eta^2} = -\frac{1}{2} \left(\frac{dZ}{d\eta} \right)^2 = -\frac{\mathcal{F}^2}{8\Phi^2}. \quad (\text{B.1b})$$

Neglecting \mathcal{B} reduces the order of the system, and so we suppress the boundary conditions on the highest-order quantities (12b,e) and (22c). (B.1) is, therefore, to be solved subject to

$$2\Phi'(1) - (1 + \nu)\Phi(1) = 1 - \nu, \quad Z(1) = 0, \quad (\text{B.2a,b})$$

with:

$$Z(\mathcal{R}^2) = -d, \quad \mathcal{R}\Phi'(\mathcal{R}^2) - \frac{\Phi(\mathcal{R}^2)}{\mathcal{R}} = 0, \quad (\text{B.3a,b})$$

for a cylindrical indenter; or

$$Z(\mathcal{R}^2) = -d + \frac{\mathcal{R}^2}{2\mathcal{R}_s}, \quad Z'(\mathcal{R}^2) = \frac{1}{2\mathcal{R}_s}, \quad \mathcal{R}\Phi'(\mathcal{R}^2) - \frac{\Phi(\mathcal{R}^2)}{\mathcal{R}} = -\frac{\mathcal{R}^3}{16\mathcal{R}_s^2}, \quad (\text{B.4a-c})$$

for a spherical indenter.

Two integrations of (B.1b) leads to

$$(A\Phi)^{1/2}(1 + A\Phi)^{1/2} - \sinh^{-1} \sqrt{A\Phi} = \frac{\mathcal{F}A^{3/2}}{2}\eta + B, \quad (\text{B.5})$$

for integration constants A and B , while (B.1a) leads to

$$Z = \frac{2}{\sqrt{A}} \sinh^{-1} \sqrt{A\Phi} - C, \quad (\text{B.6})$$

for some constant C .

Applying the boundary conditions (B.2)–(B.4) leads to a system for the unknown constants A , B , and C . To simplify these equations, we define $\Phi_0 := A\Phi(\mathcal{R}^2)$ and $\Phi_1 := A\Phi(1)$; the system then takes the form

$$\Phi_1^{1/2}(1 + \Phi_1)^{1/2} - \sinh^{-1} \Phi_1^{1/2} = \frac{\mathcal{F}A^{3/2}}{2} + B, \quad (\text{B.7a})$$

$$\Phi_0^{1/2}(1 + \Phi_0)^{1/2} - \sinh^{-1} \Phi_0^{1/2} = \frac{\mathcal{F}A^{3/2}}{2}\mathcal{R}^2 + B, \quad (\text{B.7b})$$

$$\mathcal{F}A^{3/2} \left(1 + \frac{1}{\Phi_1} \right)^{1/2} - (1 + \nu)\Phi_1 = (1 - \nu)A, \quad (\text{B.7c})$$

$$\frac{2}{A^{1/2}} \sinh^{-1} \Phi_1^{1/2} = C, \quad (\text{B.7d})$$

with:

$$\mathcal{F}A^{3/2} \left(1 + \frac{1}{\Phi_0}\right)^{1/2} = \frac{2\Phi_0}{\mathcal{R}^2}, \quad (\text{B.8a})$$

$$\frac{2}{A^{1/2}} \sinh^{-1} \Phi_0^{1/2} = C - d, \quad (\text{B.8b})$$

for a cylindrical indenter; or

$$\mathcal{F}A^{3/2} \left(1 + \frac{1}{\Phi_0}\right)^{1/2} = \frac{2\Phi_0}{\mathcal{R}^2} - \frac{\mathcal{R}^2 A}{8\mathcal{R}_s^2}, \quad (\text{B.9a})$$

$$\frac{2}{A^{1/2}} \sinh^{-1} \Phi_0^{1/2} = C - d + \frac{\mathcal{R}^2}{2\mathcal{R}_s}, \quad (\text{B.9b})$$

$$\Phi_0 = A\mathcal{F}\mathcal{R}_s, \quad (\text{B.9c})$$

for a spherical indenter. Eliminating B and C from system (B.7) with (B.8) or (B.9), leaves a force–displacement equation given implicitly by parameters Φ_0 , Φ_1 , and A (with \mathcal{R} as an extra unknown in the spherical case); the resulting system for a cylindrical indenter is given in (16).

We now consider the two limits $\mathcal{F} \rightarrow 0$ and $\mathcal{F} \rightarrow \infty$ to derive explicit asymptotic force–displacement relations; in particular, equation (B.7c) determines how A and Φ_1 relate to the applied force, \mathcal{F} .

Appendix B.1. Moderate indentation forces ($\mathcal{F} \rightarrow 0$)

In the small indentation limit, $\mathcal{F} \rightarrow 0$, eq. (B.7c) gives $A \sim \Phi_1 = 4/\mathcal{F}^2 + \mathcal{O}(1)$, which leads to the parametric force–displacement relationship for a cylindrical indenter:

$$\frac{d}{\mathcal{F}} = \log \frac{4}{\mathcal{F}} - \sinh^{-1} \Phi_0^{1/2} + \mathcal{O}(\mathcal{F}^2), \quad (\text{B.10a})$$

where Φ_0 solves

$$\frac{\Phi_0^{3/2}}{(1 + \Phi_0)^{1/2}} \sim \frac{4\mathcal{R}^2}{\mathcal{F}^2}. \quad (\text{B.10b})$$

For a spherical indenter, we find

$$\frac{d}{\mathcal{F}} = \log \frac{4}{\mathcal{F}} - \sinh^{-1} \Phi_0^{1/2} + \frac{\mathcal{R}^2}{2\mathcal{R}_s\mathcal{F}} + \mathcal{O}(\mathcal{F}^2), \quad (\text{B.11a})$$

where Φ_0 and \mathcal{R} are given by

$$\Phi_0 \sim \frac{4\mathcal{R}_s}{\mathcal{F}}, \quad (\text{B.11b})$$

$$\frac{\mathcal{R}^2}{2\mathcal{R}_s\mathcal{F}} \sim \Phi_0^{1/2} \left\{ (2 + \Phi_0)^{1/2} - (1 + \Phi_0)^{1/2} \right\}. \quad (\text{B.11c})$$

Taking the limits $\mathcal{F} \ll \mathcal{R} < 1$ and $\mathcal{R} \ll \mathcal{F} \ll 1$ leaves the leading-order solutions (18) for a cylindrical indenter and (24) for a spherical-capped indenter.

Appendix B.2. Large indentation forces ($\mathcal{F} \rightarrow \infty$)

In the large indentation limit, $\mathcal{F} \rightarrow \infty$, eq. (B.7c) gives $A = \mathcal{O}(\mathcal{F}^{-2/3})$ and $\Phi_1 = \mathcal{O}(1)$; inserting these into the relevant system and rearranging leads to the parametric force–displacement relation for a cylindrical indenter:

$$\frac{d}{\mathcal{F}^{1/3}} = \frac{2}{(1+\nu)^{1/3}} \frac{\sinh^{-1} \Phi_1^{1/2} - \sinh^{-1} \Phi_0^{1/2}}{\Phi_1^{1/2} (1+\Phi_1)^{-1/6}} + \mathcal{O}(\mathcal{F}^{-2/3}), \quad (\text{B.12a})$$

where Φ_1 and Φ_0 are given by

$$\frac{1+\nu}{2} \frac{\Phi_1^{3/2}}{(1+\Phi_1)^{1/2}} \sim \frac{[\Phi^{1/2}(1+\Phi)^{1/2} - \sinh^{-1} \Phi^{1/2}]_{\Phi_0}^{\Phi_1}}{1 - \mathcal{R}^2}, \quad (\text{B.12b})$$

$$\frac{1+\nu}{2} \frac{\Phi_1^{3/2}}{(1+\Phi_1)^{1/2}} \sim \mathcal{R}^{-2} \frac{\Phi_0^{3/2}}{(1+\Phi_0)^{1/2}}. \quad (\text{B.12c})$$

For a spherical indenter, we find

$$\frac{d}{\mathcal{F}^{1/3}} = \frac{2}{(1+\nu)^{1/3}} \frac{\sinh^{-1} \Phi_1^{1/2} - \sinh^{-1} \Phi_0^{1/2}}{\Phi_1^{1/2} (1+\Phi_1)^{-1/6}} + \frac{\mathcal{R}^2}{2\mathcal{R}_s \mathcal{F}^{1/3}} + \mathcal{O}(\mathcal{F}^{-2/3}), \quad (\text{B.13a})$$

where Φ_1 , Φ_0 , and \mathcal{R} are given by

$$\frac{1+\nu}{2} \frac{\Phi_1^{3/2}}{(1+\Phi_1)^{1/2}} \sim \frac{[\Phi^{1/2}(1+\Phi)^{1/2} - \sinh^{-1} \Phi^{1/2}]_{\Phi_0}^{\Phi_1}}{1 - \mathcal{R}^2}, \quad (\text{B.13b})$$

$$\frac{1+\nu}{2} \frac{\Phi_1^{3/2}}{(1+\Phi_1)^{1/2}} \sim \frac{\Phi_0^{3/2}}{(\mathcal{R}_s^3 \mathcal{F})^{1/2}}, \quad (\text{B.13c})$$

$$\frac{\mathcal{R}^2}{4(\mathcal{R}_s^3 \mathcal{F})^{1/2}} \sim (2 + \Phi_0)^{1/2} - (1 + \Phi_0)^{1/2}. \quad (\text{B.13d})$$

An asymptotic expansion in $\Phi_0 \rightarrow 0$ (this is equivalent to taking $\mathcal{R} \rightarrow 0$) leads to the leading-order solutions (21) for a cylindrical indenter and (25) for a spherical-capped indenter. [Note that although $\Phi_0 = \Phi_1 = 0$ is a solution of (B.12b,c), this does not correspond to the limit of large indentation depth. A local expansion around $\Phi_0 = \Phi_1 = 0$ shows that this is not the correct solution unless $\mathcal{R}^2 = (3\nu - 1)/(\nu + 1)$, as discussed by Vella and Davidovitch (2017) for the case of a point indenter ($\mathcal{R} = 0$); they noted that the trivial solution is only viable if $\nu = 1/3$. Instead, equations (B.12b) and (B.13b) must be divided through by $\Phi_1^{3/2}$.]

Appendix C. Choice of strain energy density function

The construction of a strain energy density function, $W(\lambda_1, \lambda_2, \lambda_3)$, is a difficult process, being dependent on both the material and experiment at hand (see discussion in *e.g.* Holzapfel 2002, Chp. 6). Here, our focus is on a qualitative representation of hyperelasticity, and so we seek suitable strain energy density functions. In this Appendix, we construct the strain energy function used in this paper.

We require our stresses to be consistent with Hookean elasticity in the small strain limit, *i.e.* $T_\xi \sim \sigma_{rr}$ and $T_\phi \sim \sigma_{\theta\theta}$ in the limit $\lambda_\xi, \lambda_\phi \rightarrow 1$, for T_ξ and T_ϕ defined in (29). Taylor expanding (29) around $\lambda_\xi = \lambda_\phi = 1$ (see *e.g.* Horgan and Saccomandi 2004), provides the consistency conditions

$$W_{I_1} + W_{I_2} = -(W_{I_2} + W_{I_3}) = \frac{\mu}{2}, \quad (\text{C.1a})$$

$$W_{I_1 I_1} + 4W_{I_1 I_2} + 2W_{I_1 I_3} + 4W_{I_2 I_2} + 4W_{I_2 I_3} + W_{I_3 I_3} = \frac{\lambda}{4} + \frac{\mu}{2}, \quad (\text{C.1b})$$

for Lamé parameters μ and λ , and tensor invariants

$$I_1 := \lambda_1^2 + \lambda_2^2 + \lambda_3^2, \quad I_2 := \lambda_1^2 \lambda_2^2 + \lambda_3^2 \lambda_1^2 + \lambda_2^2 \lambda_3^2, \quad I_3 := \lambda_1^2 \lambda_2^2 \lambda_3^2, \quad (\text{C.2a-c})$$

where we define

$$W_{I_i} := \left. \frac{\partial W}{\partial I_i} \right|_{\lambda_1=\lambda_2=\lambda_3=1} \quad \text{and} \quad W_{I_i I_j} := \left. \frac{\partial^2 W}{\partial I_i \partial I_j} \right|_{\lambda_1=\lambda_2=\lambda_3=1}. \quad (\text{C.3a,b})$$

(Provided (C.1) holds, our membrane equations (30) linearize to the zero bending-stiffness Föppl-von Kármán equations, (7) with $B = 0$, independent of the choice of strain energy density.)

As discussed in the main text, we concentrate on neo-Hookean and Gent solids. To formulate their strain density functions, it is convenient to split the strain energy into an isochoric and volumetric part:

$$W(\lambda_1, \lambda_2, \lambda_3) \equiv W_{\text{iso}}(I_1, I_2, I_3) + W_{\text{vol}}(J), \quad (\text{C.4})$$

for the tensor invariants (C.2) and Jacobian $J = I_3^{1/2}$. The benefit of this is that the isochoric part contains the full hyperelastic material model; that is,

$$\text{Neo-Hookean:} \quad W_{\text{iso}} = \frac{\mu}{2} \left[(I_1 - 3) - \log I_3 \right], \quad (\text{C.5a})$$

$$\text{Gent:} \quad W_{\text{iso}} = \frac{\mu}{2} \left[-\frac{1}{b} \log [1 - b(I_1 - 3)] - \log I_3 \right], \quad (\text{C.5b})$$

for some empirical parameter b based on the finite extensibility limit of the material ($1/b \equiv \max\{I_1 - 3\}$). Since the neo-Hookean model is recovered as $b \rightarrow 0$ in the Gent model, we consider only the Gent formulation henceforth. The volumetric part is a requirement of compressible materials ($J \neq 1$) and can take many constitutive forms (Holzapfel, 2002; Horgan and Saccomandi, 2004). We take

$$W_{\text{vol}} = c_1 \log J + c_2 (\log J)^2 + c_3 (J^2 - 1), \quad (\text{C.6})$$

for constants c_i ; the c_i are determined by inserting the volumetric strain energies (C.6) with Gent isometric strain (C.5b) into the consistency relations (C.1) to give:

$$c_1 = -2c_3 = c_2 + b\mu - \frac{\lambda}{2}, \quad (\text{C.7})$$

where we are free to choose c_2 . Without loss of generality, we choose $c_2 = 0$ (to suppress the $(\log J)^2$ term) — this form was first proposed by Simo and Miehe (1992) in the context of thermoplasticity. Coupling with (C.5b), our strain energy density function takes the final form

$$W = \frac{\mu}{2} \left[-\frac{1}{b} \log [1 - b(I_1 - 3)] + (\beta - b)(I_3 - 1) - (1 + \beta - b) \log I_3 \right], \quad (\text{C.8})$$

for $\beta := \lambda/2\mu = \nu/(1 - 2\nu)$.

Although (C.8) is not necessarily the simplest choice of strain energy function, it does contain some nice properties for our work. These include: compressibility (*i.e.* a variable Poisson’s ratio ν), which is needed to match up with the general Föppl-von Kármán equations; a variable nonlinearity (through the finite-extensibility parameter b), giving us the ability to continuously vary our hyperelastic material model and include the common neo-Hookean hyperelasticity as a sub-case ($b = 0$); the finite-extensibility property of Gent ($b > 0$) could qualitatively model properties seen in complex materials such as graphene and MoS₂ (specifically the finite length of bonds between atoms).

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