A Bulk Perturbation in an Alexander Brush

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ABSTRACT: We consider the effects of a small, intruding object in the bulk of a flat Alexander polymer brush in a melt state. A pointlike object creates a pressure field that has a quadrupolar form at short distances but decays exponentially away from the source. The induced surface deformation also decays exponentially away from the source, and in both cases, the decay length is of the order of the brush thickness. We also discuss the interaction between two immersed objects.

1. Introduction

Grafted polymer brushes are systems formed by irreversibly attaching the chain ends of polymers to a solid surface. Increasing the density of attachments forces the polymer chains to stretch outward from the surface, thus forming a layer with a brushlike structure. These systems are accessible to experimental probes such as force microscopy and neutron scattering. Many technological applications of these systems are possible, as a brush coating can radically modify the surface properties of a solid object. To date, however, no complete theoretical understanding exists of these systems and the characteristics of their response to external forces. The article of Halperin *et al.*[1] provides an overview of the general aspects of brushes and related microstructures.

The particular brush model we will deal with is the Alexander brush, which assumes that all the chains stretch from the bottom to the top of the layer. The assumed stretching is very large, as the thickness of the brush is taken to be much larger than the radius of gyration of a normal state chain. Furthermore, we consider only the case of a melt condition, in which the bulk of the layer is incompressible.

The Alexander approximation has been used in theoretical works to try to infer properties of brushes in general. We should note that, while it is in principle possible to have systems in which all the chains are stretched up to the top of the layer, the assumption of very strong stretching is not true in the physical systems currently available. The use of these approximations, however, makes the system amenable to theoretical study.[2,3] Furthermore, the Alexander model has been particularly useful in the study of the long-wavelength properties of the system, i.e., for deformations with characteristic length λ that is large compared to the thickness of the layer, h.

We focus on the problem of describing the interaction of the brush with a small object that has penetrated its bulk. To describe the bulk of the brush, we use here a formalism developed in a previous paper.[4] The polymer chains are described by nonintersecting average paths, and a free energy is written in terms of such paths and is then analyzed by means of a variational principle. The bulk insertion appears within this formalism as a small modification of the usual constraints that the system satisfies. A perturbation theory calculated is then possible, using the volume of the object as the perturbation parameter. We will find the pressure induced by the object in the surrounding bulk as well as the top surface deformation. The deformation is sketched qualitatively in Figure 1.

A previous treatment of this problem by similar methods was presented by Williams and Pincus.[5] The same form of the free energy was employed, but the chain ends played no role in their considerations so that the effects induced by the free surface of the brush were nil. As we shall see, however, boundary effects cause the pressure and deformation fields to decay exponentially away from the insertion. We expect this important feature to be present in the actual physical system.

The extra pressure field that results from one insertion is readily interpreted as the strength of the interaction between two localized insertions into the bulk of the brush. Our final result will be that the nature of the interaction depends on the relative positions of the insertions.

2. The Free Energy

We begin by considering a reference state (without perturbation) invariant under translations in directions tangent to the base surface. In this state, the top surface is also flat, the thickness of the brush is h, and the grafting density σ (chains/area) is taken to be constant. The chains are monodisperse with monomer number N, and they are equally stretched with average paths perpendicular to the base surface. Given the melt condition, the volume per monomer v is constant across the layer, thus giving us the relation $h = \sigma v N$.

We follow the deformation of each chain and specify the position of a given monomer as a function of its body coordinates. First, the coordinates for the physical space $\mathbf{X} = (x,y,z)$ are taken such that the base surface is described by $z = 0$ and the top surface by $z = h$. The body coordinates are defined to coincide with the physical space coordinates in the reference state, so that a monomer $\mathbf{r} = (r,s,t)$ belongs to a chain attached at the point $\mathbf{x}_0 = (r,s,0)$ and its monomer number along that chain is $n = (t/h)N$. With this notation, the average path of a chain can be written as a function of one parameter, $\mathbf{X}(t) = \mathbf{X}(\mathbf{x}_0,t)$. The use of body coordinates greatly simplifies the treatment of the boundary conditions of the problem.

The main contribution to the free energy of the system is of entropic origin. When a chain has a stretched average path, the number of configurations accessible to it is greatly reduced. One can express this contribution in terms of a phenomenological constant a that

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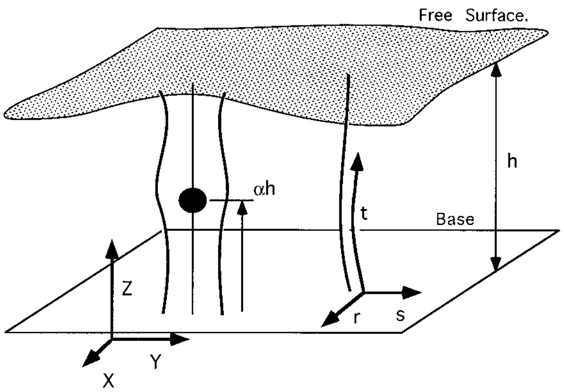

Figure 1. Schematic of a section of the brush. The Alexander condition is assumed, and the insertion is shown modifying the chain trajectories and the free surface shape. The space coordinate system (x,y,z) and body coordinate system (r,s,t) are shown. The brush height h and insertion height αh are indicated.

weighs the local stretching of the chains (measured in units of length) as

$$F = \sum_{\text{all chains}} \int \mathrm{d}t \, \frac{ah}{2N} \dot{\mathbf{X}}^2 \qquad (1)$$

where the overdot indicates partial derivation with respect to t. This density is integrated over the monomer number of each chain. We remark that this functional measures only the free energy difference with respect to a free melt.

It is also important to include a surface energy term to stabilize the system at small wavelengths. With our choice of coordinates, area of the deformed top surface can be expressed in terms of the deformation field $\mathbf{X}$ and its derivatives with respect to the body coordinates evaluated at the $(t = h)$ plane. The surface tension will be denoted as γ, and then

$$F = \gamma \int \mathrm{d}S \qquad (2)$$

The melt condition imposes an important restriction on the allowed configurations for the chains. Since we have assumed their trajectories to be nonintersecting, the map from body to space coordinates is one to one, so that the Jacobian of this mapping has to be constant all over the layer. In the variational approach, this can be enforced by adding a Lagrangian-multiplier $P(\mathbf{r})$, coupled to the constraint

$$J(\mathbf{X};\mathbf{r}) - 1 \equiv \det(\partial X_i/\partial r_j) - 1 = 0 \qquad (3)$$

A small fluctuation of this expression away from zero corresponds to the occupation of a volume $V + \mathrm{d}V$ by a group of monomers originally inside a smaller volume V. Since the multiplier penalizes this deformation by $P\,\mathrm{d}V$, we identify P with the pressure inside the layer.

At this point, we can set up the free energy to consider the inclusion of a small object in the layer. If the small intruding volume $V = \epsilon h^3$ is concentrated in the neighborhood of monomers $\mathbf{r} + \Delta\mathbf{r}$, the physical volume associated with these monomers will change as

$$\Delta r \Delta s \Delta t \rightarrow (1 + \epsilon)\Delta r \Delta s \Delta t \qquad (4)$$

thus modifying the required value of the Jacobian to

$$J = 1 + \epsilon h^3 \delta(\mathbf{r} - \mathbf{r}_0) \qquad (5)$$

In general, we can consider the inclusion of a small continuous distribution of matter with volume fraction $\epsilon\rho(\mathbf{r})$. In this case, the constraint on the Jacobian reads

$$J(\mathbf{r}) - (1 + \epsilon\rho) = 0 \qquad (6)$$

Having the density ρ depend on the body coordinates rather than the physical-space coordinates facilitates later considerations, and the differences induced in the quantities calculated appear only in higher orders in perturbation theory.

Detailed evaluation of the properties of the system that follows from the proposed free energy is presented in the Appendix. The main results are explained in the following sections.

3. Bulk Equilibrium in the Presence of a Perturbation

Solution of the equilibrium equations for the brush before the insertion of the external object shows that the presence field P_0 is uniform across the layer and has a value of $P_0 = b = a\sigma v$. We also obtain the free energy density per unit area for this state:

$$f_0 = \frac{1}{2}bh + \gamma = \frac{1}{2}a\sigma^3 v^2 N + \gamma. \qquad (7)$$

A very important result regarding the unperturbed state is the fact that a finite surface tension is required to make it stable. In the following, we restrict our analysis to the stable region, given by

$$\gamma \geq 0.056bh. \qquad (8)$$

The origin and nature of the instability of the Alexander brush have been discussed in a previous work.[4]

As shown in the Appendix, once the perturbation (with prescribed density ρ) is inserted, the main equations that describe deviations from the ground state are

$$b\ddot{\mathbf{X}}_1 = \nabla P_1 \qquad (9)$$

$$\nabla \cdot \mathbf{X}_1 = \rho \qquad (10)$$

where a double overdot indicates a second derivative with respect to the t coordinate, ∇ is the gradient operator with respect to the body coordinates, and the subscripts indicate the order of the quantity in perturbation theory. These equations lead to our main result:

$$\nabla^2 P_1 = b\ddot{\rho} \qquad (11)$$

These constitutive equations coupled with proper boundary conditions completely determine the equilibrium state of the brush.

The boundary conditions are specified as follows: In the base of the brush, no displacements are allowed. In the free surface, we require the equilibrium of forces that act on a chain segment lying in the surface. The respective equations are presented in full in the Appendix.

A comment on the peculiar form of the source is in order. First, let us note an electrostatic analogy to eq 11. The electrostatic potential for a point charge obeys a similar equation, with a source term proportional to a delta function. If the source of the potential is a dipole or a higher multipole, the source term in the equation can be written in terms of derivatives of delta functions. In our case, if we model the density ρ of a concentrated intrusion as a delta function, the full source term, after the application of the derivatives, is equivalent to an electric quadrupole source. The origin of this quadrupole coupling can be understood in terms of the presence of a preferred axis t and the fact that the stretching does not distinguish between the two orientations along the axis. (If there were a preferred direction along the axis, we could have expected a dipole field.)

This result can also be expressed in terms of the hydrodynamic analogy of Williams and Pincus.[5] They noted that the chain shape in the neighborhood of an insertion is analogous to the flow lines of an ideal fluid passing by a stationary (spherical) object. The perturbation in the velocity field $\mathbf{v}$ created by the object is well known to have a dipolar form. In our notation, this velocity field is essentially $\dot{\mathbf{X}}$. The potential field P for the "acceleration" $\ddot{\mathbf{X}}$ is then that of a quadrupole source.

4. Pressure Response

In the Appendix, we calculate the pressure response of the brush to a density-wave insertion placed at a height αh ($\alpha < 1$):

$$\rho = h\delta(t - \alpha h) \sin(\mathbf{q}\cdot\mathbf{x}/h) \tag{12}$$

This response can be written in the form

$$P_1 = p(q,t,\alpha) \sin(\mathbf{q}\cdot\mathbf{x}/h) \tag{13}$$

where the coefficient $p(q,t,\alpha)$ is referred to as the pressure susceptibility. Given the pressure response to this source, we can recover, by superposition, the pressure field created by a general perturbation. The explicit formula for the susceptibility can be written in the form $p = p_s + p_h$, where p_s is a term associated with the source and p_h is a homogeneous contribution. Our choice for the source term is

$$p_s = h\delta(t - \alpha h) + q\Theta(t - \alpha h) \sinh[q(-\alpha + t/h)] \tag{14}$$

so that the homogeneous part is

$$p_h = [A(q) \cosh(qt/h) + B(q) \sinh(qt/h)]/D(q) \tag{15}$$

with coefficients and denominator

$$A(q) = -q[\sinh(q - \alpha q) + 2 \sinh(\alpha q) - gq \cosh(q - \alpha q)] \tag{16}$$

$$B(q) = -q[(4 + gq^2) \cosh(q - \alpha q) - 2 \cosh(\alpha q) - q \sinh(q - \alpha q)] \tag{17}$$

$$D(q) = -4 + 5 \cosh(q) - q \sinh(q) + gq(-\sinh(q) + q \cosh(q)) \tag{18}$$

In these expressions $g = \gamma/bh$.

The solution for the insertion of a concentrated object at height αh is obtained through

$$P_1(R,t) = \frac{b}{2\pi}\int_0^\infty q \, dq \, J_0(qR/h)p(q,t,\alpha) \tag{19}$$

in which we have introduced a cylindrical radial coordinate $R^2 = r^2 + s^2$ and J_0 is the $0th$-order Bessel function.

Starting from this formal expression, we can analyze the asymptotic behavior of the pressure. When the distance from the source is large, $R/h \rightarrow \infty$, we look at the behavior of the integrand in the Hankel transform (eq 19) at low q. From the fact that $p(q,t,\alpha)$ is analytic in q and even under inversion of q, a result from the theory of asymptotic expansions of integrals[10] shows that the total pressure must decay exponentially away from the source. The same results holds true if we look at perturbations that originate from a line source in the t direction instead of a point source.

We may further examine the long-distance behavior by analyzing the analytic structure of the pressure field averaged over the thickness of the layer, which we denote by $\langle p(q,\alpha)\rangle$. The poles of this function $\hat{q}_i$ in the upper half of the complex q plane give rise in real space to contributions proportional to $K_0(i\hat{q}_iR/h)$, where K_0 is a modified Bessel function of the second kind. For large arguments, the Bessel function K_0 decays exponentially with a decay length l proportional to the inverse of the imaginary part of the pole $l = h/\mathrm{Im}(\hat{q}_i)$ and is modulated by oscillations with a wavelength given by the inverse of the absolute value of the real part of the pole $\lambda = 2\pi h/|\mathrm{Re}(\hat{q}_i)|$. Thus, at large distances in real space, the response is dominated by the pole or poles with the smallest imaginary part.

The poles of the averaged pressure $\langle p(q)\rangle$ are the zeroes of the denominator $D(q)$, eq 18. The relative amplitude of the contribution of each pole depends on the height of the insertion α, but the location of the poles is a function of the surface tension parameter g only. The poles are located symmetrically with respect to the $\mathrm{Im}(q)$ and $\mathrm{Re}(q)$ axes, so that one has only to look at the upper right quadrant of the complex plane. In this region we find that, for positive surface tensions, there is an infinite number of poles located in the imaginary axis and at most one complex or two real poles.

For the purpose of establishing the asymptotic behavior of the averaged pressure, we should consider first the real and complex poles. For g positive but smaller than the critical value $g_c = 0.056$, there are two real positive poles and the system is unstable as explained in a previous work.[4] Above this value, the poles become complex and acquire a rapidly increasing imaginary part, making possible large oscillations in the response for g near the critical value g_c. This is the dominant pole in the stable region only for values of g such that $0.056 < g < 0.057$, where its imaginary part is smaller than that of all poles in the imaginary axis. This pole eventually merges into the imaginary axis, where it becomes one more of the imaginary poles. The path followed by this pole is shown in Figure 2a.

The next pole to consider is the one that starts at $g = 0$ as the imaginary pole that is closest to the real axis. The path followed by this pole is shown in Figure 2b. Just slightly above the critical value g_c, this pole becomes dominant and up to a value of $g = 1.8$ stays in the imaginary axis, providing a decay length $l \approx h$. Above this value of g, this pole becomes complex and with its counterpart in the left half-plane produces an exponential decay modulated by an oscillation. At large values of the surface tension, this pole flows toward the origin.

In the case of fairly large surface tension, $g \gg 1$, the free surface deformations are softened and the excluded monomers are displaced mostly laterally. This requires a pressure field that decays more slowly with the transverse distance. More precisely, we can write the leading contributions to the average pressure in this regime as

$$\langle p(q)\rangle \approx 1 + \frac{3q^2}{q^4 + 3/g} \tag{20}$$

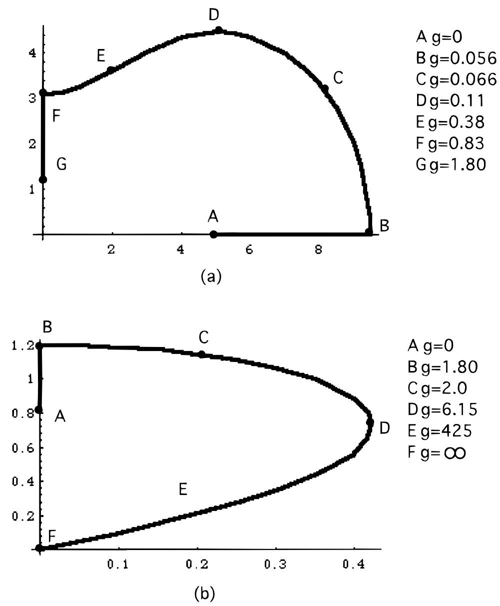

Figure 2. Location of the most important poles of the averaged pressure response $\langle p(q)\rangle$ as the surface tension parameter g changes: (a) shows the path of the pole that starts at a real value and (b) shows the path of the pole that starts as the first pole in the imaginary axis.

This expression develops a singularity $\sim 1/q^2$, as $g \to \infty$, thus implying a logarithmic decay of the pressure field when we force the top surface to remain flat. At large but finite values of g, the dominant poles are q $(\pm 1 + i)(3/g)^{1/4}$. The decay length and the oscillation wavelength (up to a factor) are then

$$l_p = \left(\frac{g}{3}\right)^{1/4} h \qquad (21)$$

We have, on the other hand, that the short-distance behavior of the pressure field is dominated by the quadrupole field. For a pointlike source with effective volume ϵV, this is

$$P_1(R,t) = \epsilon b V[R^2 - 2(t - \alpha h)^2]/[R^2 + (t - \alpha h)]^{5/2} \quad (22)$$

Thus, the precise form of the pressure field interpolates between this short field form and the far away behavior described above.

We should also comment on the fact that there is not a strong dependence of the pressure on the insertion height αh. For example, in the limits $\alpha = 0$, $\alpha = 1$, one finds a well-behaved pressure field and surface deformations with properties not different from those described above. We expect these limits to correspond to situations similar to those considered before by Fredrickson *et al.*[3] and Solis and Pickett[4] in which the pressure field and internal deformations are calculated when the free surface of the brush is held in a deformed shape.

Once the solution for the pressure field of a small object is available, it is easy to calculate the interaction energy between two such objects with volumes ϵV_1 and ϵV_2. This is simply

$$F_{int} = \epsilon^2 V_2 P_1 = \epsilon^2 V_1 P_2 \qquad (23)$$

This expression follows easily from the perturbative expansion of the free energy under the assumption that both objects are small. The pressure field P_1 is the one created by the first object, evaluated at the region of introduction of the second.

We note that according to the short distances expression for the pressure field (eq 22), the pressure field is negative in regions right above and below the source ($R \approx 0$). This means that the interaction between two insertions is attractive when they are aligned vertically. If their relative separation is horizontal, the induced pressure field felt by a second insertion is positive and their interaction is thus repulsive. The interaction is, however, negligible when the distance between insertions is larger than the decay length.

The repulsion between insertions separated by a horizontal distance arises from the squeezing (and thus stretching) of chains between them. The attraction between vertically separated insertions indicates that the deformations created by a single insertion are softened by similar deformations below or above it. Further, it is easy to see that vertical alignment avoids the squeezing of chains. We can generalize this result to state that it is easier to insert structures that are compatible with the strong vertical alignment of the brush.

A gold−copolymer system with characteristics similar to those described by our model was recently studied by Morkved *et al.*[7] We can estimate the interaction between gold spheres in diblock lamellae observed in their experiment. The lamellae are symmetric diblocks of polystyrene and poly(methyl methacrylate) containing 300 monomers per block. They were observed to have a layer thickness h of about 10 nm at a temperature of 430 K. In these conditions, $P_0 h^3 \approx 10^2 kT$. The gold spheres had diameters $\approx$ 4 nm. If the spheres were separated by a distance of twice their diameters, the estimated interaction energy would be $F_{int} \approx 10^{-1} kT$. This is a somewhat small effect, particularly for such large spheres. The available diblocks, however, do not present a case of strong stretching. If strongly stretched brushes are constructed, the effects can become more important.

5. Surface Deformations

We are also interested in the deformations induced by an intrusion in the brush and in particular in the deformations of the free surface of the brush. Direct observations of such deformations can be a useful experimental technique for the study of the brushes.

We define the surface susceptibility $\chi(q,\alpha)$ of the layer to be the ratio of the amplitude of the surface deformation $z_1(t{=}h) = h_1 \cos(\mathbf{q}\cdot\mathbf{x}/h)$ to the amplitude of the previously considered source $\rho = h\delta(t - \alpha h) \sin(\mathbf{q}\cdot\mathbf{x}/h)$. The analytical expression for this quantity is found in the Appendix to be

$$\chi(q,\alpha) = [2 \cosh((1 - \alpha)q) - \cosh(\alpha q) + q \sinh(\alpha q)]/D(q) \quad (24)$$

with $D(q)$ as in eq 18.

The surface susceptibility tends to unity as $q \to 0$. This is an expected result since the introduction of a uniform slab of material within the layer displaces the surface uniformly by the thickness of the insertion.

Figure 3 shows the surface susceptibility for a source position $\alpha = 1/2$ for three different values of the surface tension: right above the stability threshold, for a moderate value $g = 1$, and for a rather large tension $g = 20$.

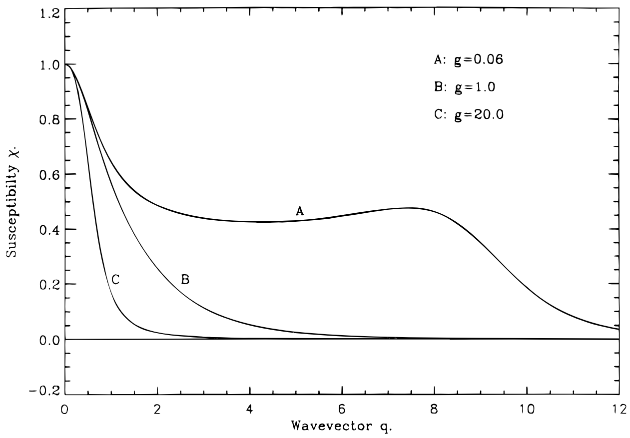

Figure 3. Surface susceptibility plotted for three different values of the surface tension. The external matter wave has been inserted at midheight $\alpha = {}^1/_2$.

As with the pressure, the form of the surface susceptibility implies an exponential decay away from the source. Its asymptotic behavior follows that of the averaged pressure discussed before, but with one important difference. We observe that in the presence of a surface tension $g \gg 1$ the susceptibility will be strongly suppressed for all nonzero wave vectors by a factor of g^{-1}. Although we still have the poles near the origin discussed before, the amplitude associated with them cancels their effects and the response has no singularities.

6. Conclusions

We have found that a pointlike object inserted in an Alexander brush can be described as a quadrupolar source for the pressure field in the bulk of the brush. Also important is the fact that the boundary conditions of the problem lead to an exponential decay for the induced fields, with a decay length comparable to the height of the layer.

We have also discussed the orientational; structure of the interaction between two immersed objects. The brush presents a smaller resistance to insertions that least disrupt the original orientation of the chains.

A more precise calculation of the pressure and deformation fields for general brushes has to be made by dropping the Alexander approximation. Work in progress,[8] in the framework of Semenov's self-consistent field approach,[9] indicates that many of the qualitative features found by using the Alexander approximation are preserved in this more accurate description. In the self-consistent field approach, the structure of the near pressure field does not present an attractive region above and below the insertion, but it is still true that an elongated object is better placed parallel to the original orientation. Another preserved feature is that boundary conditions also induce screening in the pressure field and the surface deformation with a decay length that is also on the order of the layer's thickness. At the quantitative level, however, the contrast of the results from both approaches indicates that, unless the system to be studied does physically realize the Alexander condition, the results obtained cannot be generalized to non-Alexander brushes.

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Appendix

This appendix provides the details of the perturbation theory and variational principle calculations required to obtain our main results.

First, we present the equilibrium equations associated with our free energy functional. To zeroth order in perturbation theory, we will recover the flat reference state. To first order, we obtain the linear response of the system.

Gathering together all the free energy contributions that were presented in section 2, we have

$$F = \int \mathrm{d}r\,\mathrm{d}s \int_0^h \mathrm{d}t \Big\{ \frac{1}{2} b \dot{\mathbf{X}}^2 - P(\mathbf{r})(J(\mathbf{X};\mathbf{u}) - 1 - \epsilon\rho) + \delta(t-h)\gamma[(\partial_r \mathbf{X})^2 (\partial_s \mathbf{X})^2 - (\partial_r \mathbf{X}\cdot\partial_s \mathbf{X})^2]^{1/2} \Big\} \quad (25)$$

where $b = av\sigma^2$ and the area of the deformed surface has been written in terms of its parametrization by the base coordinates using a standard formula from differential geometry.[11]

The calculations are simpler if we consider only states with translational symmetry in one direction tangential to the base surface, say r. Thus, with the exception of $X(\mathbf{u}) = r$, all other fields are independent of r. The free energy functional then simplifies to

$$F = \int \mathrm{d}s \int_0^h \mathrm{d}t \Big[\frac{1}{2} b(\dot{Y}^2 + \dot{Z}^2) - P(Y'\dot{Z} - Z'\dot{Y} - 1 - \epsilon\rho) + \delta(t-h)\gamma(Y'^2 + Z'^2)^{1/2} \Big] \quad (26)$$

where the primes denote partial differentiation with respect to s.

For a functional $S = \int_V L(\phi,\partial_\mu\phi)$, we have the well-known Euler−Lagrange equations $\partial_\mu(\partial L/\partial(\partial_\mu\phi)) - (\partial L/\partial\phi) = 0$ to be satisfied in the interior of the region V. When no restrictions are put on the boundaries, the boundary terms also have to be extremes.[10] This condition yields the not so frequently used natural boundary conditions $n_\mu(\partial L/\partial(\partial_\mu\phi)) = 0$, where n_μ is the unit vector normal to the surface S bonding V. Such boundary conditions are easily interpreted as the requirement that no external force acts at the boundary of the system.

The Euler−Lagrange equilibrium equations that follow from our functional are

$$-b\ddot{Y} + P'\dot{Z} - \dot{P}Z' = 0 \quad (27)$$

$$-b\ddot{Z} - P'\dot{Y} + \dot{P}Y' = 0 \quad (28)$$

$$Y'\dot{Z} - Z'\dot{Y} = 1 - \epsilon\rho \quad (29)$$

The boundary conditions for the fields at the base of the brush are obtained simply by the requirements of constant grafting density and no deformation of the base surface:

$$Y'(t=0) = 1 \quad (30)$$

$$Z(t=0) = 0 \quad (31)$$

At the top of the layer, on the other hand, we use the force-free boundary conditions:

$$\left[b\dot{Y} + PZ' - \gamma\left(\frac{Y'}{\sqrt{Y'^{\,2} + Z'^{\,2}}}\right)'\right]_{t=h} = 0 \qquad (32)$$

$$\left[b\dot{Z} - P\dot{Y} - \gamma\left(\frac{Z'}{\sqrt{Y'^{\,2} + Z'^{\,2}}}\right)'\right]_{t=h} = 0 \qquad (33)$$

where all the quantities are evaluated at $t = h$.

It is easy to check that our reference state (*i.e.*, the flat brush without perturbation) is indeed a solution of the previous equations, and in doing so we learn that the pressure P_0 takes the constant value $b = av\sigma^2$. In addition, a simple evaluation gives us the free energy per unit area in this state (eq 7).

We now expand all fields and equations in powers of ϵ around the reference state. The linear response to a perturbation with arbitrary density distribution can be obtained by superposition of densities of the form

$$\rho = h\delta(t - \alpha h)\sin(qs/h) \qquad (34)$$

where αh is the height at which the insertion is placed. We can consider all other fields having the same, definite wavenumber:

$$\mathbf{X} = \mathbf{X}_0 + \epsilon\mathbf{X}_1 + ... = \mathbf{r} + \epsilon\mathbf{X}_1 + \epsilon^2\mathbf{X}_2 + ... \qquad (35)$$

$$Y = Y_0 + \epsilon Y_1 + ... = s + \epsilon h\, y(t)\cos(qs/h) + ... \qquad (36)$$

$$Z = Z_0 + \epsilon Z_1 + ... = t + \epsilon h\, z(t)\sin(qs/h) + ... \qquad (37)$$

$$P = P_0 + \epsilon P_1 + ... = b + \epsilon b\, p(t)\sin(qs/h) + ... \qquad (38)$$

The linearization of the equilibrium conditions (eqs 27−29) reads

$$b\ddot{\mathbf{X}}_1 = \nabla P_1 \qquad (39)$$

$$\nabla\cdot\mathbf{X}_1 = \rho \qquad (40)$$

By taking t derivatives in eq 40, we may eliminate $\mathbf{X}_1$ from both (39) and (40) to obtain

$$\nabla^2 P_1 = b\ddot{\rho} \qquad (41)$$

That is, the pressure disturbance field is harmonic except in the region occupied by the source. The linearized boundary conditions are

$$[\dot{Y}_1 + Z_1']_{t=h} = 0 \qquad (42)$$

$$[\dot{Z}_1 - Y_1' - ghZ_1'']_{t=h} = 0 \qquad (43)$$

$$[Z_1]t_{=0} = 0 \qquad (44)$$

$$[Y_1']_{t=0} = 0 \qquad (45)$$

In the second of these equations, we have introduced the dimensionless constant $g = \gamma/bh$.

We now solve our equations with a source as in eq 34. For this, we rewrite all fields in terms of the pressure:

$$h\dot{z} = p - p(0) \qquad (46)$$

$$z = (h/q^2)[\dot{p} - \dot{p}(0)] - p(0)t \qquad (47)$$

$$y = [p - p(0)]/q \qquad (48)$$

We look for a solution for the pressure field in the form $p = p_s + p_h$, p_s being a term associated with the source and p_h a homogeneous part. The result is eqs 14−18.

The deformation of the free surface is evaluated immediately as

$$h_1 = z(t{=}h) \qquad (49)$$

$$h_1 = [2\cosh((1-\alpha)q) - \cosh(\alpha q) + q\sinh(\alpha q)]/D(q) \qquad (50)$$

To recover the solution for a concentrated object from the solution for a density wave, se use for the source

$$\rho = h^3\delta(t - \alpha h)\delta(r)\delta(s) = \frac{h^3}{2\pi R}\delta(R)\delta(t - \alpha h) \qquad (51)$$

in which we have introduced a radial coordinate $R^2 = r^2 + s^2$. Since the source can be written as the superposition

$$\rho(R,t) = \frac{h}{4\pi^2}\int \mathrm{d}k\,\mathrm{d}l\; e^{i(kr+ls)/h}\delta(t - \alpha h) = \frac{h}{2\pi}\int_0^\infty q\,\mathrm{d}q\; J_0(qR/h)\delta(t - \alpha h) \qquad (52)$$

where J_0 is the zeroth-order Bessel function, the solution for the pressure field can be written as the superposition

$$P_1(R,t) = \frac{b}{2\pi}\int_0^\infty q\,\mathrm{d}q\; J_0(qR/h)p(q,t,\alpha) \qquad (53)$$

with $p(q,t)$ being the solution for a plane wave of wave vector q. Similarly, the deformation of the top surface shape is found to be

$$h_1(R) = \frac{h}{2\pi}\int_0^\infty q\,\mathrm{d}q\; J_0(qR/h)\chi(q,\alpha) \qquad (54)$$

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