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Simple View on Fingering Instability of Debonding Soft Elastic Adhesives

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We study the crack-front fingering instability of an elastic adhesive tape that is peeled off a solid substrate. Our analysis is based on an energy approach using fracture mechanics and scaling laws and provides simple physical explanations for (i) the fact that the wavelength depends only on the thickness of the adhesive film and (ii) the threshold of the instability, and (iii) additionally estimates the characteristic size of the fingers. The scaling laws for these three observables are in agreement with existing experimental data.

Introduction

Thin viscous, elastic, and viscoelastic films occur widely in industrial processes and play a major role in the current manufacturing of nanodevices. (Visco-)elastic films are often used as adhesives,[1] and their behavior, for example, under peeling[2] and tensile loading,[3] has thus been the subject of a large amount of work.[4−7] From a fundamental point of view, a major issue concerns instabilities during tensile deformation in confined geometries (see, e.g., ref 8 and references therein). Recently, it has been shown that when a flexible plate is peeled off a layer of purely elastic adhesive bound to a rigid substrate, fingerlike debonding patterns form along the crack-front.[9,10] For these elastic materials, in contrast to the classical viscous Saffman−Taylor instability,[11] no transport of matter is involved. Experiments also show that the wavelength of the instability does not depend on the rate of peeling and scales linearly with the film thickness h.[9] The crack-front instability disappears if the film thickness exceeds a critical thickness h_c proportional to $(D/E)^{1/3}$, where D is the flexural rigidity of the flexible plate and E is the elastic modulus of the film,[12] implying that the confinement plays a major role in this instability.

The wavelength dependence of the finger pattern was first explained by investigating a slightly different geometry, where the elastic film is brought close to a contactor.[9,13] Recently, also the peeling geometry was studied in detail by performing a linear stability analysis of the straight debonding front within the full elastic equations.[14,15] The wavelength dependence and also the threshold thickness h_c could be obtained approximately. However, these studies are involved, since the finger pattern is no longer a plane problem and the boundary conditions are stepwise functions, and thus, they rely on numerical evaluations and asymptotic analysis.

In this work, we thus propose simple arguments, based on fracture mechanics and scaling laws, to describe the relevant features of the fingering instability. (As usual for a scaling law description, numerical prefactors in our results are only rough estimates.) The simplicity of the description allows one to increase the comprehension of the problem: it explains why the wavelength of the fingering instability only depends on the film thickness while it can strongly vary in the cavitation geometry. More importantly, it offers a simple picture to understand the appearance of a critical film thickness above which the fingers disappear and gives a prediction for the length of the fingers, that was not yet available from calculations using the full elastic equations. The geometry we consider is an adhesive tape, composed of a thin incompressible elastic film firmly bound to a flexible plate, that is peeled off a flat rigid substrate.[2] We first discuss the elastic debonding instability in a cross section perpendicular to the peeling direction, a situation related to the classical Griffith problem[16] of rupture in solids, but under confinement. Then we generalize this picture to account for the full geometry of the fingers and discuss their disappearance upon an increase of the film thickness, and their typical length. To validate subsequent assumptions, for the physical parameters, we use the following typical values (taken for PDMS films as studied in ref 17): elastic modulus of the adhesive, $E \sim 10^6$ N·m^{-2}; work of adhesion, $W_0 \sim 10^{-2}$ J·m^{-2}; typical film thickness of the adhesive, $h \sim 10^{-4}$; typical plate rigidity of the upper plate ("tape"), $D \sim 0.01-1$ N·m.

Confined Griffith Problem

In a first step, we look at the debonding instability associated with the finger formation in a cross section perpendicular to the peeling direction. In this section, taken to be the yz-plane, see Figure 1, fingers correspond to two-dimensional "bubbles" (see right part of Figure 2) of equal size and regularly spaced with

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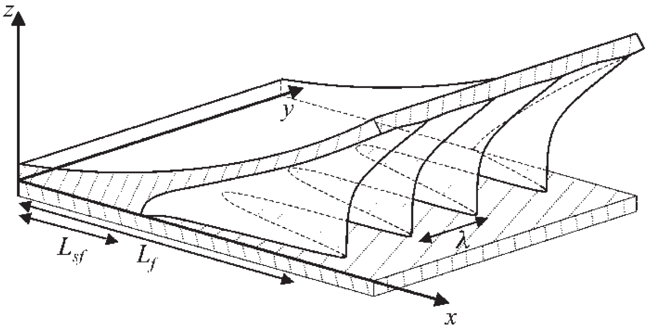

Figure 1. Schematic of the finger patterned adhesive zone upon peeling an elastic adhesive tape (modeled as an elastic film with a rigid plate on top) off a substrate. For $x < 0$, the tape is assumed to be flat. The fingers start a distance L_{sf} away from $x = 0$ and extend up to a length L_f. The fingers formed have a typical wavelength λ (in y-direction).

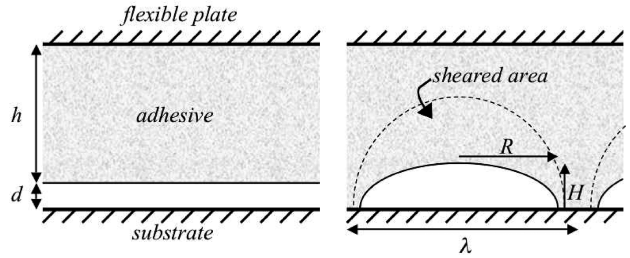

Figure 2. Two possible scenarios for an infinite elastic film lifted from a flat substrate by a small distance d: (left) the whole film detaches from the substrate; (right) the film stays partially in contact with the substrate, forming equally spaced bubbles of equal size.

wavelength λ in the y-direction. Similar scaling theories on bubble formation upon loading have been discussed in the literature.[18,19] We will assume here that we are in a perfect adhesive tape geometry; that is, the elastomer is permanently attached to the upper plate and debonds only from the lower plate. We thus neglect the respective surface energies at the upper contact.

Consider first the incompressible elastomer to be stretched in z-direction by a length $d \ll h$. If the elastomer stays completely attached, the energy per unit surface is $f_a = \gamma_{FS} + E(d/h)^2h$, where the first term is the film−substrate interface energy and the second is the elastic energy stored (modulus E). If instead the lower interface debonds completely, cf. the left part of Figure 2, the energy of this "completely detached" state is simply $f_d = \gamma_{FA} + \gamma_{SA}$, since now both the film and the substrate are in contact with air (with energies γ_{FA} and γ_{SA}, respectively), while the elastic energy is completely released. One finds $f_d < f_a$ if

$$d > d_c = \sqrt{\frac{W_0 h}{E}} \tag{1}$$

with $W_0 = \gamma_{FA} + \gamma_{SA} - \gamma_{FS}$ being the work of adhesion. Expectedly, the system debonds if the separation of the two plates is sufficiently large so that the elastic energy exceeds the work of adhesion.

Let us now look at the Griffith-like problem of bubbles appearing at the lower interface under the stress given by the displacement d. In his work on rupture in solids, Griffith[16] assumed a small disk-shaped crack of radius R inside the sample that is subject to a fixed imposed external stress σ_{ext}. Due to this stress, the energy density in the material is $e = \sigma_{ext}^2/2E$. The growth of the crack costs interface energy but allows the elastic stress to relax. By scaling, the stress goes from 0 at the free surface of the crack to σ_{ext} over a distance of the order of the size R of the crack. Thus, the gained energy through stress relaxation is roughly $(2R)^2(\sigma_{ext}^2/2E)$. The total energy variation (per unit length) due to the crack is

$$\Delta E \simeq 2\gamma(2R) - (2R)^2\frac{\sigma_{ext}^2}{2E} = 4\left(\gamma R - \frac{R^2\sigma_{ext}^2}{2E}\right) \tag{2}$$

This gives the well-known critical radius for crack growth,

$$R_c \simeq \frac{2E\gamma}{\sigma_{ext}^2} \tag{3}$$

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that is, for $R > R_c$ the crack will open, while for $R < R_c$ it will "heal". In our case, to create new surfaces, it costs the work of adhesion, W_0, rather than 2γ and the stress due to the initial stretching by d is $\sigma_{ext} \simeq E(d/h)$. This yields $R > R_c = W_0h^2/Ed^2$. Let us assume that at the lower interface bubbles are free to form, that is there exists a "precrack" with $R > R_c$. While the Griffith argument is for an infinite system and constant stress, in our case the growth of the cavity relaxes substantially the external stress and when the size of the cavity reaches the thickness of the adhesive film, $R = h$, the relaxation of stress has reached the upper surface and the bubble growth will stop there. Thus, localized bubbles grow if $h \geq R \geq R_c = (W_0h/Ed^2)h$. Obviously, this is only possible if again $d > d_c$. Thus, the following interpretation is at hand: as soon as there are precracks of large enough size (but smaller than h), the film will partially detach via formation of cavities. Since the thresholds of complete and partial debonding are the same, this will proceed until the film is completely detached.

Although in the situation described above bubbles will grow until complete debonding of the film, fingers (corresponding in the yz cross section to periodic bubbles) might still be favorable compared to a straight debonding front. The reason is that, for a periodic state, the incompressibility can be adjusted inside the cross section, while for a straight front, to fulfill the incompressibility, the system has to create shear in the x-direction. Fully developed bubbles will have the radius $R \simeq h$ and will have fully relaxed the stress over their size. Thus, the energy of a periodic bubble state with a wavelength of λ is

$$f_P = \frac{2R}{\lambda}(\gamma_{FA} + \gamma_{SA}) + \left(1 - \frac{2R}{\lambda}\right)\left(\gamma_{FS} + E\frac{d^2}{h}\right)_{|R=h} \tag{4}$$

The first term is the part of the film where the cavity is (size $2R$ per wavelength λ) and comprises only the energy of the two interfaces with air since the elastic stress is completely relaxed. The other term is the still attached region (size $\lambda - 2R$ per λ), where the interface energy is γ_{FS} and the film is still stressed with $\sigma \simeq E(d/h)$. Clearly, this is just an interpolation between f_a (for $R = 0$) and f_d (for $\lambda = 2R$). To discuss the effect of incompressibility for the periodic state, we compare the energy relaxed through creation of a cavity with the energy needed to create the same cavity by distorting the nonstressed elastomer, which should be of the same order:

$$E\left(\frac{d}{h}\right)^2 h \simeq E\left(\frac{H}{2R}\right)^2 R \tag{5}$$

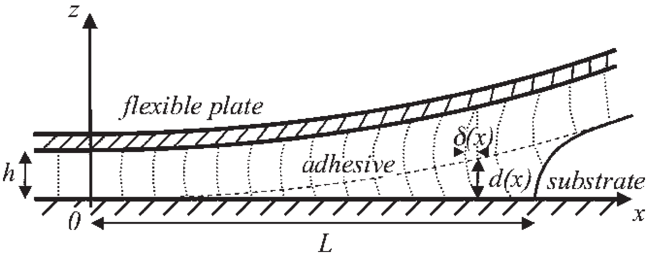

Figure 3. Side view of the physical situation investigated. The adhesive tape is peeled off a flat rigid substrate. The dotted lines sketch the deformation field in the soft adhesive joint.

Here, H is the height of the cavity. Incompressibility imposes the relation $H(2R) \simeq \lambda d$ which is simple conservation of volume, cf. Figure 2. Using this relation for H, we get from eq 5: $d^2/h = \lambda^2 d^2/(16R^3)$ which together with $R \simeq h$ yields

$$\lambda \simeq 4h \tag{6}$$

This result is in good agreement with experiments, reporting values close to $\lambda = 4h$,[12] as well as $\lambda = 2.3h$,[8] depending on the specific geometry. Since we use scaling laws, however, the numerical prefactor found here is approximate and the good agreement is rather by accident. More refined theories yield $\lambda = 3.4h$[14] and $\lambda = 3.3h$.[15] The emphasis here should be laid on the fact that the wavelength does not depend on any physical parameter other than h, and on the simple interpretation of the intervention of the film thickness h via the maximum bubble size R.

Using $R = h$ and $\lambda = 4h$ in eq 4, for the energy per length of the periodic, state we get

$$f_{\mathrm{p}} = \frac{1}{2} W_0 + \gamma_{\mathrm{FS}} + \frac{1}{2} E \frac{d^2}{h} \tag{7}$$

Fingering versus Straight Crack Front

To proceed from the previous analysis to the fingering instability, we now have to take into account the third dimension, that is, the direction of peeling along the x-axis; see Figure 3. The curvature of the flexible backing of the film leads to variations of the distance d along the fingers. We assume that the flexible backing, of flexural rigidity D, is much more rigid than the elastic film, so that one can neglect the curvature energy of the film as compared to the one of the plate. This is the same as stating that for the nondimensional parameter

$$K = \frac{D}{Eh^3} \tag{8}$$

$K \gg 1$ holds. (In this framework, $\alpha = K^{1/3}$ is sometimes called the confinement parameter.) Indeed, using $D \sim 0.01 - 1\ \mathrm{N \cdot m}$ as well as $E \sim 10^6\ \mathrm{N \cdot m^{-2}}$ and $h \sim 10^{-4}$m as given in the introduction, one gets $K \sim 10^4 - 10^6$. Ghatak et al.[17] have shown that the vertical displacement of the plate and its curvature are slightly nonmonotonous functions of x. For simplicity, and for it should not change the scaling laws, we assume that the plate is perfectly flat in the $x < 0$ region and that it has a constant curvature ζ in the $x > 0$ region. Then one can use the well-known scaling[20,21] for the equilibrium curvature of a plate that is peeled off a substrate,

$$\zeta \simeq \left(\frac{W_0}{D}\right)^{1/2} \tag{9}$$

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as a function of work of adhesion and flexural rigidity. With no adhesive film in between, the state just described, that is, flat for $x < 0$ and constant curvature ζ for $x > 0$, would be the equilibrium state. However, in the presence of the adhesive film, an adhesive bridge, cf. Figure 3, will be present since the adhesive can gain energy by being attached for $x > 0$ provided that the cost of elastic energy is smaller. We thus will have to compare the energy of the finger pattern and the energy of a straight front with a reference state ("no adhesive") that is completely detached for $x > 0$.

All along the adhesive zone, with the curvature of the flexible backing given by eq 9, the distance d between the substrate and the peeled-off film is a function of x and given simply by $d(x) \simeq \zeta x^2/2$. If $d(x)$ varies slowly enough ($x \ll \zeta^{-1}$, $\lambda \ll \zeta^{-1}$), we can write for the energy $f(d,x) = f(d(x))$. Since $\zeta \simeq 0.1 - 1\ \mathrm{m}^{-1}$ for the given parameters, this approximation is clearly justified for the resulting lengths of the adhesive zone.

For the total energy of a finger pattern of length L we get from eq 7

$$F_{\mathrm{p}}(L) = \int_0^L (f_{\mathrm{p}} - f_{\mathrm{d}})\, \mathrm{d}x = -\frac{1}{2} W_0 L + \frac{E\zeta^2}{8h} \int_0^L x^4\, \mathrm{d}x \tag{10}$$

(We do not consider the case of a varying wavelength along the adhesive zone, which would certainly have a high energy cost since it is rarely observed. Thus, we fix $\lambda = 4h$; see eqs 6 and 7.) Minimization, $\partial F_{\mathrm{p}}/\partial L = 0$, leads to an optimum length of (omitting prefactors of order one)

$$L_{\mathrm{f}} \simeq hK^{1/4} \tag{11}$$

The maximum strain at the front of the fingers is $d(L_{\mathrm{f}})/h \simeq (W_0/Eh)^{1/2}$ so that $d(L_{\mathrm{f}}) \simeq (W_0 h/E)^{1/2} = d_{\mathrm{c}}$ as in the Griffith argument. Since $(W_0/Eh)^{1/2} \sim 10^{-2}$, the assumption $d \ll h$ remains valid. Also, L_{f} is much larger than h for $K \gg 1$. The length of the fingers found, $L_{\mathrm{f}} = (Dh/E)^{1/4}$, is the same as that found by estimating the lateral width of the stressed zone within the film,[9] which is the only other estimate at hand. Experimental measurements of L_{f} on several films have shown quasi-linear dependence on $(D/E)^{1/3}$, independent of film thickness. We believe that these measurements could possibly be fitted with our result as well, since eq 11 displays only a very weak dependence ($h^{1/4}$) on thickness that is almost not noticeable for the range of experimentally investigated thicknesses, and $(D/E)^{1/3}$ is also close to $(D/E)^{1/4}$ for the experimental values. The length of the fingers is independent of the work of adhesion, W_0, since the elastic energy scales like the deflection of the plate squared, that is, linear in W_0 at equilibrium, as does the surface energy. This is clearly a result of the assumed linearity of the elasticity and might not be the case for strong deformations of the adhesive.

Let us now consider the simplest possible mode of the adhesive zone, that is, a straight crack front with a deformation field invariant along the y-axis. In the $x > 0$ region, as in the finger case, the backing is characterized by the curvature ζ and $d(x) \simeq \zeta x^2/2$ between the backing and the substrate. The soft joint can either come off the substrate or deform to maximize its surface of contact with the substrate. We again assume that $d(x) \ll h$ and that the size L_{s} of the adhesive zone is much larger than h, that is, strong confinement, and much smaller than ζ^{-1}. As a result, the incompressibility of the adhesive imposes a strong shear which dominates the stretch. The incompressibility condition, $\partial u/\partial x + \partial w/\partial z = 0$, under confinement leads with the typical scales δ for the horizontal displacement u, cf. Figure 3, and d for the

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vertical displacement w and h for the typical dimension in the z-direction to a horizontal displacement of order $\delta(x) \simeq x\, d(x)/h$. Thus, the main contribution to shear, $\gamma(x) = \partial u/\partial z$, reads $\gamma(x) = \delta(x)/h \simeq (\zeta/2)(x^3/h^2)$. (The other contribution, $\partial w/\partial x \propto d/x \propto x$ is negligible. Normal strain is $\propto d/h \propto x^2$, as in the finger case. It is thus also smaller than the contribution from $\partial u/\partial z$ and will be neglected here.) In contrast, for the fingers, the incompressibility could already be roughly fulfilled in the yz-plane, namely, by choosing H of the bubble to be $H(2R) \simeq \lambda d$, so that $\partial u/\partial x \simeq \delta/x \simeq 0$ and thus also $\gamma \simeq 0$. However, since the finger geometry is not plane strain, this is clearly an approximation and idealization. For the straight front, the elastic energy evaluates to $hE\gamma(x)^2$ and the total energy of a straight adhesive zone of length L thus reads

$$F_s(L) = \int_0^L (f_s - f_d)\, dx = -W_0 L + \frac{E\zeta^2}{4h^3}\int_0^L x^6\, dx \qquad (12)$$

Comparing eqs 10 and 12, in the finger case, we gain less surface energy, $-W_0/2$ compared to $-W_0$, than in the straight case while the elastic penalty is higher due to shear in the straight case. In both cases, there is an optimal length, and minimization of eq 12 leads to

$$L_s \simeq hK^{1/6} \qquad (13)$$

The maximum strain for the straight front is $\gamma(L_s) = \zeta L_s^3/h^2 \simeq (W_0/Eh)^{1/2}$, that is, of the same order as in the case of the fingers. Also, $\zeta^{-1} \gg L_s$ holds as well as $L_s \gg h$, provided that $K \gg 1$.

One can easily check that $F_p(L_f) < 0$ and $F_s(L_s) < 0$; that is, both states are preferable with respect to staying completely detached for $x > 0$. More importantly, the total energy of the fingers (per unit of length in y-direction) is smaller than the total energy of the straight adhesive zone: one gets $F_p(L_f) \simeq -W_0 hK^{1/4} < -W_0 hK^{1/6} \simeq F_s(L_s)$ provided that $K \gg 1$, which confirms that finger patterns can appear. Formation of fingers allows one to avoid the strong confinement due to incompressibility.

Now we are also able to give a simple interpretation of the threshold for the fingers' appearance: In the limit $K \to 1$, the length L_f for the fingers and the length L_s for the straight front will *collapse* and the fingers will thus disappear. Looking at the definition of K, eq 8, this limit is equivalent to $h \to (D/E)^{1/3}$. This explains in a simple and intuitive fashion the occurrence of the threshold, that is, that for thicknesses $h > h_c$ no fingers can form. Moreover, it shows that the threshold scales like

$$h_c \simeq (D/E)^{1/3} \qquad (14)$$

This scaling of h_c agrees exactly with the experimental one.[12]

Two further comments on our description are in order here: first, one should note that, for very small values of x, the shear deformation of the straight front yields a lower energy than the periodic one of the fingers, $(E\zeta^2/8h)x^4 < (E\zeta^2/4h^3)x^6$, namely, if $x < L_{sf} \simeq h$. Therefore, the fingers do not begin exactly at $x = 0$ but at a distance $x = L_{sf}$, while the finger tips are located at the distance $x = L_f$ (see Figure 1). This means that the adhesive zone contains a region of high shear at the substrate level ($x < L_{sf}$), which can give birth to slippage, and the finger region with no shear on the substrate ($L_{sf} < x < L_f$). This might explain the occurrence of slippage sometimes observed upon the disappearance of the fingers. Second, next to the adhesive zone ($x \gtrsim L_s$), there should be the fracture tip, or healing zone of size h. We did not include it in the total energy of the system, since it does not depend on the length L. However, to be more precise, the fact that the stress $\sigma(L_s) \simeq (EW_0/h)^{1/2}$ is much smaller than the stress W_0/a (where a is the molecular distance characterizing the surface forces) required to initiate a fracture leads to the presence of this healing zone where the stress scales like $(EW_0/(L_s - x))^{1/2}$.

Conclusions

In this work, we provided very simple arguments for the appearance of finger patterns during debonding of an adhesive tape consisting of a rigid plate and a thin layer of soft adhesive elastomer. Our treatment is not meant to derogate much more refined theories at hand, but rather to give simple interpretations of the physical mechanisms: First, the fact that the wavelength of the finger pattern depends only on the thickness of the elastic film is due to the incompressibility of the film and to the maximum size, given by the film thickness, over which stress can relax upon "bubble" growth (a bubble being a cross section through the finger pattern). Second, we predict the length of the fingers to be of the order $L_f \simeq hK^{1/4}$ with $K = D/Eh^3$ being a nondimensional measure of the confinement. Since the length of a straight debonding front scales like $L_s \simeq hK^{1/6}$, for $K \to 1$, these two lengths collapse and the fingers disappear, giving a simple explanation for the existence of a threshold and that the critical thickness of the film scales like $h_c \simeq (D/E)^{1/3}$. The finger length has not been obtained before by more refined treatments, and the scaling $hK^{1/4}$ depends only weakly on the film thickness and thus might fit the existing experimental data. The strong confinement for $K \gg 1$ together with the incompressibility leads to an elastic penalty for the straight front due to shear and thus favors the fingers where the incompressibility can be more easily accounted for by the periodic modulation.

We should stress that we studied here only the simple case of a purely elastic film bound to the backing. Real adhesives are often viscoelastic, and the rheology can come into play as an additional complication,[22] leading, for example, to a dependence of the instability on the peeling velocity. The theory presented in this work is simple enough that it might serve as a starting point for investigating these advanced questions.

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