

Two Universal Regimes of Adhesive Film Peeling

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Abstract—A numerical model describing the process of thin adhesive film peeling off a solid surface is considered. A qualitative distinction of the “microscopic” picture from the notions of the standard theory of adhesion (based on the energy balance) consists in that separate elements of the film peel off the surface along virtually the same trajectory irrespective of the direction of the external force action. A more thorough analysis shows that a fine difference between the scenarios of behavior in the vicinity of the touch point still exist, so that one can speak of the two classes of universality corresponding to $\alpha < \pi/2$ and $\alpha > \pi/2$. The results of analysis of the proposed microscopic model allow a more correct macroscopic criterion to be formulated for the adhesive surface peeling. © 2005 Pleiades Publishing, Inc.

Introduction. In recent years, increasing attention has been devoted to the phenomenon of adhesion in various fields such as the contact mechanics, tribology, and the technology of adhesives and, especially, self-adhesive materials [1]. A large number of investigations have been devoted to theoretical aspects of the adhesive interactions as well as to their practical aspects [1–3]. One particular task of these investigations is inspired by the need of robot technology in creating artificial surfaces of the gecko foot type capable of sticking (reversibly) to solid surfaces of various kinds. Theoretical investigations of the adhesive contacts usually proceed from the main idea of the classical Johnson–Kendall–Roberts (JKR) theory [4], which is based on the analysis of a balance of the elastic energy of deformation of contacting bodies and the contact surface energy. However, as will be shown below, this simple microscopic approach does not always provide adequate description of the laws of adhesion.

The simplest example of application of the energy approach in the theory of adhesion is offered by a thin, flexible and nonstretchable film peeling off a flat solid surface (Fig. 1). The force per unit length of the peel-off line at an angle α relative to the substrate surface will be denoted $\mathbf{F} = \{F_x, F_z\} = \{F \cos \alpha, F \sin \alpha\}$, and the energy necessary to separate a unit film area from the substrate surface (i.e., the effective surface energy of interaction between the film and substrate) will be denoted γ . Let us calculate the angle α for which the peel-off line occurs in equilibrium. As the contact length between the film and the substrate surface increases by Δl , the film energy decreases by $\gamma \Delta l$, the end of the film travels over a certain distance s and performs the work Fs against the pull-off force F . In equilibrium, we have $Fs = \gamma \Delta l$. As can be readily shown, $s = \Delta l(1 - \cos \alpha)$ and, hence, $F_0(1 - \cos \alpha) = \gamma$. Here, F_0 is

the pull-off force, below which the film exhibits self-adhesion and above which the film peels off. Thus, the critical pull-off force is a monotonic function of the peeling angle:

$$F_0 = \gamma / (1 - \cos \alpha), \quad (1)$$

which decreases from $F_0 \gg \gamma$ at small angles ($\alpha \ll 1$) to $F_0 = \gamma/2$ at $\alpha = \pi$.

Microscopic model. If the above standard energy considerations were correct, it would be possible to reproduce the result described by relation (1) in a microscopic model with a surface energy modeled by interactions between solids (of the van der Waals force type). However, the results of an analysis of the microscopic model described below show that direct modeling leads to the behavior qualitatively different from that predicted by relation (1).

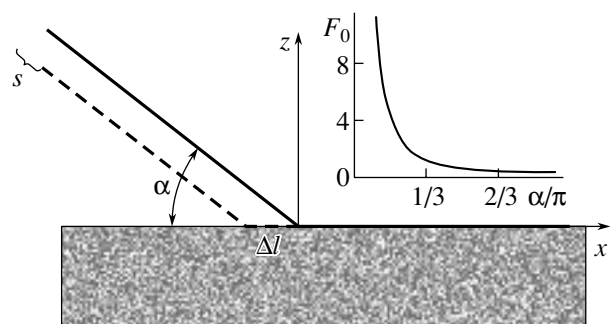


Fig. 1. Geometry of film peeling off a flat solid substrate surface (see the text for explanations). The inset shows a plot of the critical pull-off force F_0 versus angle α corresponding to formula (1).

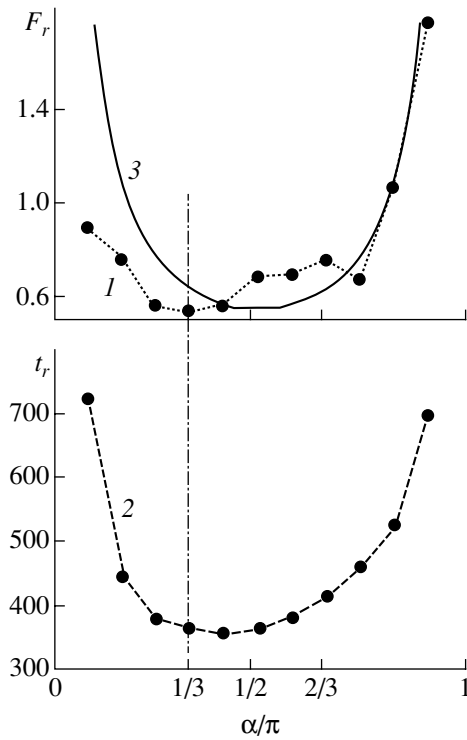


Fig. 2. Plots of (1) the critical pull-off force F_r for the film peeling and (2) the total peel-off time t_r versus angle α as determined by numerical calculations. Curve 3 corresponds to the macroscopic theory taking into account the work performed only by the vertical force component.

For the sake of simple and illustrative representation of results, the consideration below will be restricted to a one-dimensional elastic chain. However, the results of our additional numerical experiments showed that virtually all results obtained for this simple model can be reproduced in a more realistic model of a two-dimensional adhesive film. In constructing the simple microscopic model, we have to take into consideration at least the following factors: the elastic interaction between elements (segments) of the chain, their attraction to the surface of the substrate and repulsion from its deeper layers, and the external pull-off force.

In order to reproduce the elastic properties of a system, it is usually sufficient to restrict the consideration to a linear elastic bonds in a sequence of “material points” spaced by dx , which represent the neighboring surface elements with the coordinates $z(x + dx)$ and $z(x)$: $K[z(x + dx) - z(x)]$, where K is the elastic constant characterizing the interaction between mobile elements in the elastic chain. However, the linear bonds cannot determine the distance between elements in cases when the chain is freely moving in space rather than supported on a certain “substrate.” For adequate modeling of such a chain, the elastic bonds should be nonlinear [5–7]. In a widely used simple variant (see, e.g., [8] and references therein), it is possible to restrict the consideration to a fourth-order interaction potential of the

type $U^{\text{elastic}}(r) = r^2(1 - r^2/2)/2$, where $r^2 = dx^2 + [z(x + dx) - z(x)]^2$. Then, the required components $F_x^{\text{elastic}} = -U^{\text{elastic}}/x$ and $F_z^{\text{elastic}} = -U^{\text{elastic}}/z$ of the force of interaction between elements are characterized by the cubic nonlinearity that does not admit rupture of the chain and (for the elastic constant $K = 4$ used below) and quite rigidly sets the distance between elements.

The surface acts upon each element in the chain with the van der Waals force with the components $F_x^{\text{adhesion}} = -U^{\text{adhesion}}/x$ and $F_z^{\text{adhesion}} = -U^{\text{adhesion}}/z$ corresponding to the adhesion potential U^{adhesion} . This potential can in most cases be represented by a pair of Gaussians with the opposite signs and variable width, which describe the attraction partly compensated by the repulsion at short distances: $U^{\text{adhesion}}(r) = C\exp(-r^2/c) + D\exp(-r^2/d)$. This “seeding” potential (calculated for $C = 0.8$, $c = 0.2$, $D = -8$, and $d = 1$) is depicted by thick gray curve in Fig. 4. Since this potential does not provide for a repulsion of the chain from the “bulk” substrate, it has to be supplemented with a δ -like term $U^{\text{bulk}} = U_0\delta(z)$ ($U_0 \gg D$; in the calculation, $U_0 = 300$), which represents a step force supporting the sticking chain on the substrate surface: $F_z^{\text{bulk}} = -U^{\text{bulk}}/z$.

For certainty, let us consider the case of a chain peeled off the surface by the external force $\mathbf{F}^{\text{ext}} = K^{\text{ext}}(\mathbf{V}t - \mathbf{r}_1)$, which is developed by a spring extended between the first chain segment $\mathbf{r}_1 = \{x_1, z_1\}$ and a holder moving away at the angle α with the velocity $\mathbf{V} = \{V_x, V_z\} = \{V\cos\alpha, V\sin\alpha\}$ (in the calculation, $V = 0.2$).

In the overdamped limit, the dynamic equations for the model described above appear as

$$\begin{aligned} \frac{x}{t} &= F_x^{\text{elastic}} + F_x^{\text{adhesion}} + F_x^{\text{bulk}}; \\ \frac{z}{t} &= F_z^{\text{elastic}} + F_z^{\text{adhesion}} + F_z^{\text{ext}} + F_z^{\text{bulk}}. \end{aligned} \quad (2)$$

Results and discussion. The main results of numerical modeling using system (2) are presented in Fig. 2, which shows plots of the critical pull-off force F_r for the film peeling (curve 1) and the total peel-off time t_r (curve 2) as functions of the angle α . As can be seen, the obtained behavior of $F_r(\alpha)$ cannot be described using the function $\gamma/(1 - \cos\alpha)$ predicted by the macroscopic theory (see the inset in Fig. 1).

A qualitative distinction of the “microscopic” pattern from the simple geometry used to derive relation (1) consists in that each segment of the elastic chain peels off the surface along virtually the same trajectory irrespective of the direction of the external force action. At the terminal touch point, the flat surface is tangent to this trajectory.

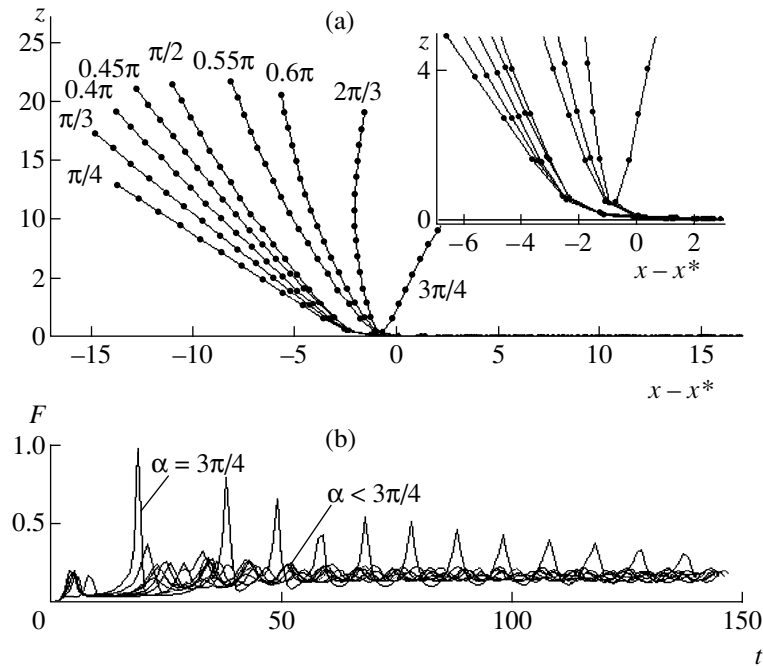


Fig. 3. Two types of the universal behavior at the point of film peeling off the substrate surface: (a) film configurations in the mobile frame related to the touch point $x = x^*$ for the peeling at various angles (the inset shows the vicinity of the touch point on a greater scale); (b) quasi-periodic time variation of the force acting on the segment at the touch point.

A more thorough analysis shows a fine difference between the scenarios of behavior in the vicinity of the touch point still exist (see Fig. 3 and the legend to this figure), so that one can speak of the two classes of universality corresponding to $\alpha < \pi/2$ and $\alpha > \pi/2$. However, this difference is much smaller that it might have been expected in the entire broad range of angles $0 < \alpha < \pi$.

As can be seen from Fig. 2, the critical force and the efficiency of work performed by this force (characterized by the total peel-off time for a chain of certain length) as functions of the angle α are almost symmetric relative to the vertical line $\alpha = \pi/2$. Taking this symmetry into account, we have to revise the process of chain peeling off the substrate surface.

Since every sequential segment peels off the surface along the tangent, the horizontal component of the external force is “spent” to perform the work (against the elastic force) on elongating the chain segments in this direction. Therefore, only the vertical force component $F_x^{\text{ext}} = F \sin \alpha$ works against the adhesion potential U^{adhesion} . Over a distance on the order of the potential width d , this force has to perform a work sufficient to detach the chain segment: $F_x d \sin \alpha = U^{\text{adhesion}}|_{z=0}$. This corresponds to the angular dependence of the pull-off force $F_r \sim 1/\sin \alpha$, which provides very good approximation (Fig. 2, curve 3) of the results of numerical modeling.

In the discrete numerical description of the system (and in real molecular chains), this scenario of segment

peeling off the surface must lead to an almost periodic step process. According to this scenario, each segment extends to a certain (nearly equilibrium) length and simultaneously peels off the surface. Subsequent extension involves the next element into this motion and so on. This process leads to a quasi-periodic time variation of the force $F(t)$ in the region of peeling (Fig. 3). As can be seen, the oscillations in $F(t)$ are most pronounced for the sliding angles close to $\alpha = \pi$. In the continuum limit (where the spacing of chain segments is much smaller than the characteristic width of the adhesion potential), $F(t)$ degenerates into a monotonic function.

The effective potential $U_{\text{eff}}^{(k)}(z)$, which acts upon an arbitrary inner k th segment in the chain at a distance z from the substrate surface, can be determined by numerical integration of the vertical force component $F^z(z(t))$ over the time series $z(t)$:

$$U_{\text{eff}}^{(k)}(z) = \int F^z(z(t)) dz(t). \quad (3)$$

Figure 4 shows the results of such integration for various angles α . The obtained potential is in good agreement (with allowance for oscillations related to the detachment of sequential discrete segments) with the “seeding” adhesion potential. For relatively small deviations of α from $\pi/2$ ($\pi/4 < \alpha < 3\pi/4$), the effective potential is virtually independent of the external force direction. This behavior confirms the above assumption that, in most cases, the chain peels off the substrate in a

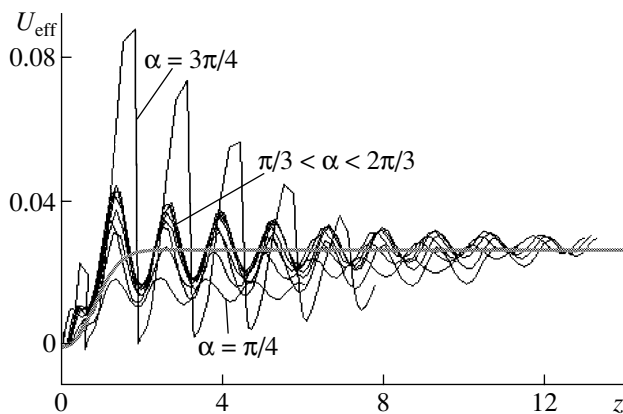


Fig. 4. The effective potential U_{eff} acting upon an arbitrary (inner) segment in the chain at a distance z from the substrate surface as determined by numerical integration of the vertical force component $F(z(t))$ over the time series $z(t)$. Thick gray solid curve shows the “seeding” adhesion potential entering into the equations of motion.

universal manner and the process rate is determined predominantly by the vertical force component.

The obtained results lead to the following conclusions.

(i) The microscopic process of adhesive bond rupture cannot be adequately described within the framework of the macroscopic approach based on the energy balance;

(ii) The most adequate macroscopic description with allowance for a real microscopic process can

employ the principle of virtual work only for the vertical force component. The horizontal component apparently performs work against other forces, such as the force of friction in the adhesive contact, which are usually not taken into consideration in the macroscopic theory of adhesion.

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SPELL: 1. gecko