Influence of Surface Roughness on Adhesion between Elastic Bodies

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We study the influence of surface roughness on the adhesion between elastic solids. We present experimental data for the force necessary to pull off rubber balls from hard rough substrates. We show that the effective adhesion (or the pull-off force) can be calculated accurately from the surface roughness power spectra obtained from the measured surface height profile.

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Adhesion between solid surfaces is a common phenomena in nature and is of extreme importance in biology and technology. Biological "applications" include cell adhesion [1–4] and the adhesive systems (for the purpose of locomotion) in insects, lizards, and some frogs [5–7]. Technological applications include pressure sensitive adhesives for envelopes and Post-it pads [8,9], as well as many high-tech applications, e.g., silicon wafer bonding [10]. However, the biggest "mystery" is not why adhesion is sometimes observed but rather why it is usually not observed [11,12]. For instance, a surface roughness of just a few nanometers is enough to remove the adhesion between clean and (elastically) hard solid surfaces [13].

Strong adhesion between two elastic solids will usually occur only if at least one of the solids is elastically soft and if both surfaces are smooth since the area of (atomic) contact between two hard solids with rough surfaces is usually only an extremely small fraction of the nominal contact area [14,15]. In addition, the solids will be elastically deformed near the contacts, and the stored elastic energy is "given back" during pull-off, thus helping to break the adhesive bond between the solids [16]. In fact, in many cases the stored elastic energy is just large enough to break the adhesive bond resulting in a vanishing pull-off force [16].

Commercial adhesives, such as pressure sensitive adhesives used for Scotch tape or Post-it pads, consist of very soft (weakly cross-linked) rubber compounds (low-frequency elastic modulus in the 10^3–10^5 Pa range), which can deform and make (almost) perfect contact even with rather rough surfaces and low squeezing pressures [17]. In addition, because rubber tack compounds usually have a nearly liquidlike long-time response, very little elastic energy will be stored at the rubber-substrate interface, resulting in a strong effective adhesion between the rubber and the substrate. On the other hand, the adhesive surface layer on the lizards' pads (and on attachment surfaces of flies, bugs, and crickets) is made from a rather stiff material (keratinlike proteins, with an elastic modulus of order 10^9 Pa) [5]. But in this case millions of years of optimization (via the principle of natural selection) has resulted in a highly structured ~ 100 μm thick layer on the pad surface, which is built in a hierarchical manner from fibers and platelike structures. This layer is elastically soft on all length scales from millimeter to nanometer, which allows the lizard to adhere and move on very rough (vertical) surfaces, such as freshly cleaved stone surfaces [6,7].

The breaking of the atomic bonds between two elastic solids during "pull-off" usually occurs by propagating an interfacial crack. For purely elastic solids, the elastic energy stored at the interface because of surface roughness will flow to the crack tip and facilitate the interfacial bond-breaking process, resulting in the small adhesion observed in most situations. However, for some materials, in particular, for rubberlike materials, a large (bulk) viscoelastic energy dissipation may occur close to the crack tip. As a result, strong effective adhesion (i.e., a large pull-off force) may be observed in situations where a purely elastic solid (with the same elastic modulus as the low-frequency elastic modulus of the viscoelastic solid) would exhibit negligible adhesion [18–20].

The standard and most well-defined method to study the adhesion between two solids is a ball-on-a-flat configuration. If at least one of the solids is elastically soft, the Johnson-Kendall-Robbets (JKR) theory can be used to analyze the experimental data. This theory predicts how the radius of the circular contact area depends on the (squeeze or pull) force F. The most important result is the pull-off force given by [21]

$$F_{\text{pull-off}} = \frac{3\pi}{2} R\Delta\gamma, \tag{1}$$

where R is the radius of the ball and $\Delta\gamma$ is the interfacial binding energy per unit surface area when two flat surfaces are brought into contact. Note that (1) does not depend on the elastic modulus of the solids.

Equation (1) is also valid for rough surfaces, assuming that the surface roughness power spectra has a roll-off wavelength λ_0 (see below), which is much smaller than the radius r_0 of the contact area between the ball and the substrate. However, in this case one must replace $\Delta\gamma$ with the effective interfacial energy γ_{eff}, which depends on the nature of the surface roughness and on the elastic modulus of the solids. For purely elastic solids one has $A_0\gamma_{\text{eff}} = A\Delta\gamma - U_{\text{el}}$. Here A_0 is the nominal contact area, A is the area of real (atomic) contact, and U_{el} is the elastic energy stored at the interface as a result of the surface roughness, i.e., U_{el} is the energy necessary to deform the rubber surface so that it makes atomic contact with the substrate over the area A.

In this Letter we present the first detailed study of adhesion, where the full surface roughness power spectra $C(q)$ is included in the analysis of the experimental data. We study the contact between rubber balls with smooth surfaces and differently prepared substrate surfaces. The substrates are elastically much harder than the rubber and can be considered as rigid. The surface morphology of the substrates are fully characterized by the surface height $h(\mathbf{x})$ measured using atomic force microscopy, from which the power spectra's $C(q)$ have been obtained using the fast Fourier transform [12].

In a classic paper Fuller and Tabor [13] have studied the influence of surface roughness on the adhesion of rubber balls to hard substrates. In order to understand the experimental data, they developed a very simple model based on the assumption of roughness on a single length scale. The surface asperities were approximated by spherical cups with equal radius of curvature, and the JKR theory was applied to each asperity contact region. This assumes that the area of real contact is much smaller than the nominal contact area, and also that the contact regions are independent of each other. Neither of these approximations is valid unless the surface roughness is so high that the effective adhesion nearly vanishes. In the present study we consider the opposite limit of complete, or nearly complete, contact where an essentially exact analysis is possible.

The substrate surfaces were prepared by vacuum evaporation of aluminum on a silicon wafer at different substrate temperatures. From the rough wafer surfaces we prepared (negative) replicas using polyvinylsiloxane. Samples for the measuring of adhesion were prepared from the polyvinylsiloxane templates using epoxy resin. We prepared 7 different surfaces that we denote by the numbers **1–7** according to increasing root-mean-square roughness. Figure 1 shows the surface topography of four of the used substrates.

The rubber lenses were prepared from polydimethylsiloxane (PDMS) silicon elastomer, which has a very low glass transition temperature, and which therefore can be considered as a nearly perfect elastic material with respect to our applications. Hemispherical cups made of glass were filled with the polymer and polymerized during 48 h at 60 °C. After polymerization, lenses were separated from the cups and the surfaces were studied for perfection with a microscope. Radii of the lenses were measured by using white light interferometer. Here we report on experiments performed at two different pull-off velocities (0.2 and 2 μm/s), for a polymer lens with radii 4.244 mm.

The pull-off force measurements were performed using a steel spring with stiffness 334.9 N/m. The PDMS lens was glued on the steel spring and mounted on a piezoelectric actuator, and the deflection of the spring was measured using a laser interferometer. The measurements were performed on a vibration isolated table. From the measured pull-off forces we calculated the effective interfacial binding energies (per unit surface area) γ_{eff} using Eq. (1).

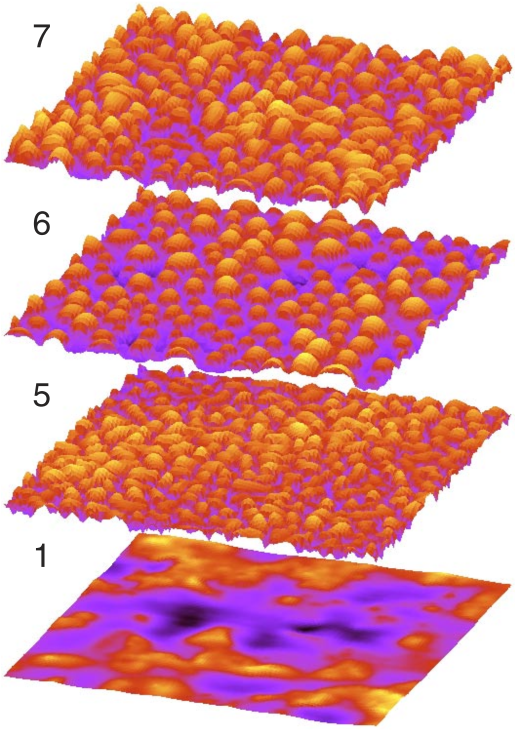

FIG. 1 (color online). The surface height profile (50×50 μm square) of surfaces **1**, **5**, **6**, and **7**, measured using the atomic force microscope.

Recently, one of us has developed a new contact mechanics and adhesion theory [14,16,22]. In this theory the surface roughness height profile $h(\mathbf{x})$ enters only via the surface roughness power spectra [12]

$$C(q) = \frac{1}{(2\pi)^2} \int d^2x \langle h(\mathbf{x}) h(\mathbf{0}) \rangle e^{i\mathbf{q}\cdot\mathbf{x}}. \tag{2}$$

Here $\langle \cdots \rangle$ stands for ensemble average, and we have assumed that the statistical properties of the surface are translational invariant and isotropic so that $C(q)$ depends only on the magnitude $q = |\mathbf{q}|$ of the wave vector $\mathbf{q}$. Figure 2 shows the surface roughness power spectra of four of the surfaces used in the present study. The curve **1** is for the smoothest surface used in our study, with the rms roughness $h_0 \approx 0.03$ μm. The surfaces shown in Fig. 1 are not self-affine fractal on any length scale [23], but this fact is not important for what follows.

The roughest surface we consider in this study has a root-mean-square roughness of about $h_0 \approx 0.2$ μm and a roll-off wave vector $q_0 \approx 10^6$ m^{-1} corresponding to the wavelength $\lambda_0 = 2\pi/q_0 \approx 10^{-5}$ m. Now, let us discuss whether the rubber-substrate adhesional interaction is strong enough to "pull" the rubber into complete contact within a "cavity" of width λ_0 and height $h_0 \ll \lambda_0$. The gain in rubber-substrate binding energy is of the order of $U_{\mathrm{ad}} \approx \lambda_0^2 \Delta\gamma$, where $\Delta\gamma$ is rubber-substrate interfacial binding energy per unit surface area for flat surfaces. However, since the adhesive interaction between the surfaces is very short ranged, the rubber must deform elastically and "bend" into the cavity to make atomic contact with the substrate surface. Since the strain in the rubber is of the order of $\sim h_0/\lambda_0$ in a volume element of order $\sim \lambda_0^3$ we get the elastic energy to be of the order of $U_{\mathrm{el}} \approx E(h_0/\lambda_0)^2 \lambda_0^3 = E h_0^2 \lambda_0$. Thus the condition $U_{\mathrm{el}} \approx U_{\mathrm{ad}}$ gives $h_0 \approx (\Delta\gamma \lambda_0/E)^{1/2}$. With $\Delta\gamma \approx 0.1\ \mathrm{J/m^2}$, $\lambda_0 \approx 10^{-5}$ m, and $E \approx 5$ MPa we get $h_0 \approx 1\ \mu$m, which is larger than the observed roughness amplitude even for the roughest surface. However, the actual situation is more complex than indicated by the calculation above since the interfacial binding energy to be used in the estimate above is not $\Delta\gamma$ deduced for perfectly smooth surfaces, but rather a smaller effective interfacial energy that takes into account the substrate roughness on length scales shorter than the roll-off wavelength λ_0.

Let us now consider the contact between an elastically soft material, e.g., rubber, with a flat smooth surface and a randomly rough hard substrate surface. Let us first assume that perfect contact occurs at the interface between the two solids. In Refs. [16,22] it has been shown that for this case the change in the free energy per unit surface area upon contact, which we denote as the effective surface energy γ_{eff}, is given by [16,22]

$$\gamma_{\mathrm{eff}} = \frac{A}{A_0} \Delta\gamma - \frac{U_{\mathrm{el}}}{A_0}, \tag{3}$$

where the increase of the surface area

$$\frac{A}{A_0} = \int_0^\infty dx (1 + x\xi^2)^{1/2} e^{-x}, \tag{4}$$

where

$$\xi^2 = 2\pi \int_{q_0}^{q_1} dq q^3 C(q), \tag{5}$$

and where the elastic energy per unit area

$$\frac{U_{\mathrm{el}}}{A_0} = \frac{\pi E}{2(1-\nu^2)} \int_{q_0}^{q_1} dq q^2 C(q). \tag{6}$$

In Eq. (3) we have assumed that the interfacial energy $\Delta\gamma$ is independent of the increase of the surface area $A - A_0$, which is likely to be a good approximation for rubber because of the high (liquidlike) mobility of the rubber chains at the rubber surface. In any case, the increase of the surface area is very small for all the studied substrates (it is highest for surfaces **3** and **4** where $A/A_0 = 1.186$) so the first term in (3) is nearly constant.

In Fig. 3 we compare the experimentally measured effective surface energy γ_{eff} for the seven different surfaces with different rms roughness amplitudes, with the theoretical calculated values, assuming perfect contact at the interface. Experimental data are shown for the pull-off velocity 0.2 (green curve **a**) and 2 μm/s (red curve **b**). The calculated results have been obtained from (3)–(6) using the measured elastic modulus of the rubber and the observed change in the interfacial energy $\Delta\gamma$ for flat surfaces. The power spectra used in the calculation were obtained directly from the height profile using (2) as described in Ref. [12]. For the substrate surface **7** the theory predicts a small negative value for $\gamma_{\mathrm{eff}}/\Delta\gamma \approx -0.119$, but in this case the noncontact state will have a lower free energy, and in the figure we have therefore used the value $\gamma_{\mathrm{eff}} = 0$ for this case.

In the present model, the physical reason for the decrease in the effective surface energy γ_{eff} is entirely due to the elastic energy stored at the interface between the solids. That is, in order for atomic contact to occur at the interface, it is necessary to deform the elastic solid, resulting in stored elastic energy at the interface. During pull-off this elastic energy is "given back" thus helping to break the adhesive bond between the solids. When $\gamma_{\mathrm{eff}} \ll \Delta\gamma$, the

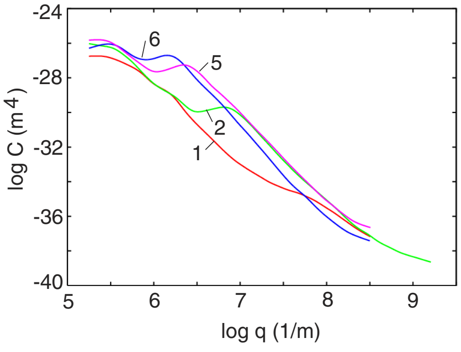

FIG. 2 (color online). The surface roughness power spectra of surfaces **1**, **2**, **5**, and **6**. The root-mean-square amplitudes of the surfaces are 0.030, 0.060, 0.168, and 0.172 μm, respectively. Curve **1** is for the smoothest surface.

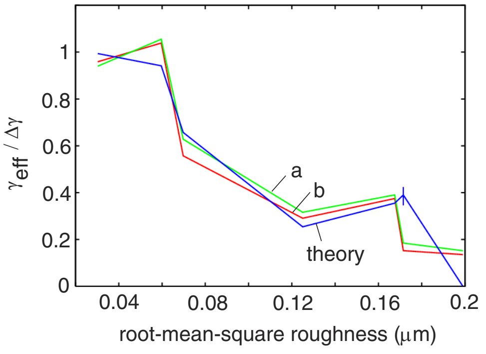

FIG. 3 (color online). The effective surface energy as a function of the root-mean-square roughness for 7 differently prepared surfaces. Blue curve: theory; green curve: **a**; and red curve: **b**. These are experimental data for the pull-off velocity 0.2 and 2 μm/s, respectively.

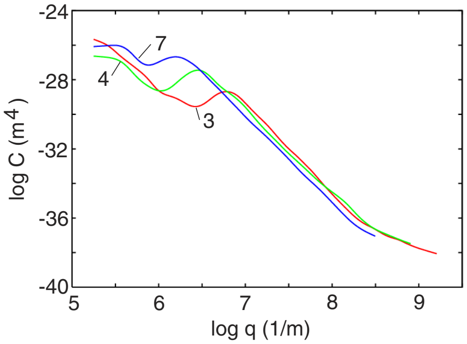

FIG. 4 (color online). The surface roughness power spectra for three rough substrates. Curve **7** is for the surface exhibiting the lowest adhesion (from theory $\gamma_{\mathrm{eff}} = 0$), while for surfaces **3** and **4**, $\gamma_{\mathrm{eff}}/\Delta\gamma = 0.657$ and 0.254, respectively. The surface area calculated from the power spectra is highest for surfaces **3** and **4** ($A/A_0 = 1.186$ in both cases) and smaller for surface **7** ($A/A_0 = 1.133$).

contact between the solids will not be complete, and in this case the treatment presented above will not be valid [16,24].

The agreement between theory and experiment in Fig. 3 is very good except for the two roughest surfaces. Thus, the theory predicts $\gamma_{\mathrm{eff}} = 0$ for surface **7**, while the experiment gives $\gamma_{\mathrm{eff}} \approx 0.135\Delta\gamma$. This may be due to the occurrence of only partial contact between the surfaces for this case. In fact, since the theory predicts a (small) negative γ_{eff}, the noncontact state has a smaller free energy than the full contact state. However, it is likely that in this case there is a partial contact state with even lower free energy, corresponding to the (small) positive value for $\gamma_{\mathrm{eff}}/\Delta\gamma$ observed in the experiment.

We note that there is no direct relation between the strength of the adhesion and the rms roughness amplitude h_0, or the roughness-induced increase in the total surface area A/A_0. To illustrate this, Fig. 4 shows the surface roughness power spectra for three of the used substrates. Curve **7** is for the surface exhibiting the lowest adhesion (from theory $\gamma_{\mathrm{eff}} = 0$), while for surfaces **3** and **4** $\gamma_{\mathrm{eff}}/\Delta\gamma = 0.657$ and 0.254, respectively. The surface area calculated from the power spectra is highest for surfaces **3** and **4** ($A/A_0 = 1.186$ in both cases), and smaller for surface **7** ($A/A_0 = 1.133$). Thus there is no correlation between the increase in the surface area and the effective adhesion. The reason for this is that the variation in γ_{eff} for the different surfaces **1**–**7** is mainly due to variations in the elastic energy (induced by the surface roughness), and this is (for the complete contact case) determined by the second moment of the surface roughness power spectra $C(q)$, while the increase in the total surface area is determined by the third moment of $C(q)$; see Eqs. (4) and (5). Similar, there is no direct relation between the root-mean-square roughness of the surfaces and the adhesional properties, since the rms roughness amplitude is determined by the first moment of $C(q)$. In fact, both the theory and the experiment show that surface **5** exhibits a larger adhesion than surface **4**, in spite of the fact that the rms roughness is considerably larger for the former surface.

To summarize, we have studied the adhesive interaction between elastic bodies with rough surfaces. We have shown how the experimental data can be understood quantitatively when the full surface roughness power spectra is used in the analysis.

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[1] J. Lee, A. Ishihara, G. Oxford, B. Johnson, and K. Jacobson, Nature (London) **400**, 382 (1999).
[2] B. N. J. Persson and F. Mugele, J. Phys. Condens. Matter **16**, R295 (2004).
[3] R. Merkel, Phys. Rep. **346**, 343 (2001).
[4] J. Israelachvili, *Intermolecular and Surface Forces* (Academic Press, London, 1992).
[5] M. Scherge and S. Gorb, *Biological Micro- and Nano-Tribology* (Springer, Berlin, 2001).
[6] B. N. J. Persson, J. Chem. Phys. **118**, 7614 (2003).
[7] B. N. J. Persson and S. Gorb, J. Chem. Phys. **119**, 11 437 (2003).
[8] K. R. Shull and C. Creton, J. Polym. Sci., Part B: Polym. Phys. **42**, 4023 (2004).
[9] C. Gay and L. Leibler, Phys. Rev. Lett. **82**, 936 (1999).
[10] C. Gui, M. Elwenspoek, N. Tas, and J. G. E. Gardeniers, J. Appl. Phys. **85**, 7448 (1999).
[11] K. Kendall, *Molecular Adhesion and its Applications* (Kluwer, New York, 2001).
[12] B. N. J. Persson, O. Albohr, U. Tartaglino, A. I. Volokitin, and E. Tosatti, J. Phys. Condens. Matter **17**, R1 (2005).
[13] K. N. G. Fuller and D. Tabor, Proc. R. Soc. A **345**, 327 (1975).
[14] B. N. J. Persson, J. Chem. Phys. **115**, 3840 (2001).
[15] R. Buzio, C. Boragno, F. Biscarini, F. B. de Mongeot, and U. Valbusa, Nat. Mater. **2**, 233 (2003).
[16] B. N. J. Persson, Eur. Phys. J. E **8**, 385 (2002).
[17] B. N. J. Persson, O. Albohr, C. Creton, and V. Peveri, J. Chem. Phys. **120**, 8779 (2004).
[18] A. D. Roberts and A. G. Thomas, Wear **33**, 45 (1975).
[19] B. N. J. Persson, O. Albohr, G. Heinrich, and H. Ueba (to be published).
[20] M. Kröger and K. Popp, Proc. Appl. Math. Mech. **4**, 99 (2004).
[21] K. L. Johnson, K. Kendall, and A. D. Roberts, Proc. R. Soc. A **324**, 301 (1971).
[22] B. N. J. Persson and E. Tosatti, J. Chem. Phys. **115**, 5597 (2001).
[23] J. Krim, I. Heyvaert, C. Van Haesendonck, and Y. Bruynseraede, Phys. Rev. Lett. **70**, 57 (1993).
[24] S. Zilberman and B. N. J. Persson, J. Chem. Phys. **118**, 6473 (2003).