Influence of surface roughness on the adhesion of elastic films

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It is shown that a self-affine roughness at the junction of an elastic film and a hard solid substrate influences considerably the adhesion of the elastic film, especially for small roughness exponents H ($H < 0.5$) and/or large long wavelength roughness ratios w/ξ with w being the rms roughness amplitude and ξ being the in-plane roughness correlation length. Analytical calculations of the local surface slope allows an estimate of the roughness effects on the adhesion energy more precisely than those presented in earlier works (especially for roughness exponents $H < 0.5$). For weak surface roughness the elastic energy contribution is significant on the film effective surface energy $\Delta \gamma_{eff}$ and on pull-off force for elastic modulus E in the range of GPa. Moreover, in the case of partial contact an estimation of the pull-off force shows that it strongly decreases with reducing contact area due to surface.

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I. INTRODUCTION

This paper addresses the question to what extent the adhesion between an elastic solid and a hard solid substrate will be affected by the surface roughness. In particular, this is a relevant question for the adhesion of polymer coatings onto metallic substrates. The topic was studied initially by Fuller and Tabor [1], and it was shown that a relatively small surface roughness could eliminate adhesion. In their model [1] a Gaussian distribution of asperity heights was considered with all asperities having the same radius of curvature. The contact force was obtained by applying the contact mechanics theory of Johnson *et al.* [2] to each individual asperity. However, this approach considers the surface roughness over a single lateral length scale. The maximum pull-off force is expressed as a function of a single parameter, which determines (the statistically averaged) a competition between compressive forces from higher asperities that try to pull the surfaces apart, and the adhesive forces from lower asperities that try to hold the surfaces together [1].

On the other hand, random rough surfaces, which are commonly encountered for solid surfaces [3,4], possess roughness over various length scales rather than over a single one. This case was considered by Persson and Tosatti [5] for the case of random self-affine rough surfaces. It was shown that when the local fractal dimension D becomes larger than 2.5, the adhesive force may vanish or at least be reduced significantly. Since $D = 3 - H$ with H being the roughness exponent, which characterizes the degree of surface irregularity (as H becomes smaller the surface becomes more irregular at short length scales), the roughness effect becomes more prominent for roughness exponents $H < 0.5$.

Nevertheless, the previous numerical studies were performed using power-law approximations for the self-affine roughness spectrum, with a sharp cutoff. This is valid for lateral roughness wavelengths $q\xi > 1$ with ξ being the in-plane roughness correlation length. This work concentrates on the effect of roughness, including contributions from roughness wavelengths $q\xi < 1$. This will be achieved by analytical calculation of the local surface slope, which subsequently determines the calculation of the adhesive interaction.

II. ADHESIVE AND ELASTIC ENERGY UNDER COMPLETE CONTACT

We assume that the substrate surface roughness is described by the single valued random roughness fluctuation function $h(\vec{r})$ with $\vec{r}$ being the in-plane position vector $\vec{r} = (x, y)$ such that $\langle h(\vec{r}) \rangle = 0$ (Fig. 1). The adhesive energy is given by

$$U_{ad} = -\Delta \gamma \int d^2 r \sqrt{1 + \vec{\nabla} h \cdot \vec{\nabla} h}. \tag{1}$$

Assuming Gaussian random roughness fluctuations [5] yields after ensemble averaging over possible random roughness configuration

$$U_{ad} = -\Delta \gamma A_{\text{flat}} \langle \sqrt{1 + \vec{\nabla} h \cdot \vec{\nabla} h} \rangle, \tag{2}$$

$$\langle \sqrt{1 + \vec{\nabla} h \cdot \vec{\nabla} h} \rangle = \int_0^{+\infty} du (\sqrt{1 + \rho^2 u}) e^{-u},$$
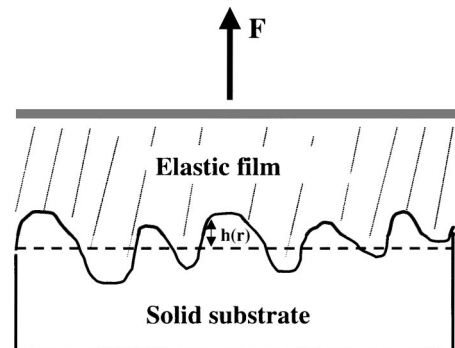

FIG. 1. Schematic of an elastic polymer film on a rough solid substrate. F is the force applied to pull off the film.

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with A_{flat} being the average macroscopic flat contact area, $\rho = \langle \sqrt{\langle (\nabla h)^2 \rangle} \rangle$ being the average local surface slope of the substrate rough surface, and $-\Delta\gamma$ being the change of the local surface energy upon contact due to elastic film or substrate interaction. Substituting in $\rho = [\langle |\vec{\nabla} h|^2 \rangle]^{1/2}$ the Fourier transform of the surface height $h(\vec{q}) = (2\pi)^{-2} \int h(\vec{r}) e^{-i\vec{q}\cdot\vec{r}} d^2\vec{r}$ with $\vec{r} = (x, y)$ being the in-plane position vector and assuming $\langle h(\vec{q}) h(\vec{q}') \rangle = \delta^2 (\vec{q}' + \vec{q}) \times \langle h(\vec{q}) h(-\vec{q}) \rangle$, i.e., translation invariance, the rms local slope ρ is given by

$$\rho^2 = \int q^2 \langle |h(\vec{q})|^2 \rangle d^2 \vec{q} = \int q^2 C(q) d^2 \vec{q}, \qquad (3)$$

where $C(q)$ is the Fourier transform of the substrate height-height correlation function $C(r) = \langle h(\vec{r}) h(0) \rangle$ that characterizes the substrate roughness. Furthermore, the elastic energy stored in the film of elastic modulus E and Poisson's ratio ν is given by

$$U_{el} = -\frac{1}{2} \int d^2 r \langle h(\vec{r}) \sigma_z(\vec{r}) \rangle \qquad (4)$$

assuming that the normal displacement field of the film equals $h(\vec{r})$. Since in Fourier space we have $h(\vec{q}) = M_{zz}(\vec{q}) \sigma_z(\vec{q})$ with $M_{zz}(\vec{q}) = -2(1-\nu^2)/Eq$ [5] and $h(\vec{q}) = (2\pi)^{-2} \int h(\vec{r}) e^{-i\vec{q}\cdot\vec{r}} d^2\vec{r}$, we obtain after substitution into Eq. (4),

$$U_{el} = A_{\text{flat}} \frac{E}{4(1-\nu^2)} \int q C(q) d^2 q. \qquad (5)$$

Notably Eq. (5) is valid for relatively weak roughness or small local surface slopes $\rho \leq \sqrt{\langle (\nabla h)^2 \rangle} < 1$.

III. SELF-AFFINE ROUGHNESS MODEL

A wide variety of surfaces or interfaces are well described by a kind of roughness associated with self-affine fractal scaling [3,4]. For self-affine surface roughness $C(q)$ scales as a power-law $C(q) \propto q^{-2-2H}$ if $q\xi \gg 1$, and $C(q) \propto$ const if $q\xi \ll 1$ [3,4]. The roughness exponent H is a measure of the degree of surface irregularity [3,4], such that small values of H characterize more jagged or irregular surfaces at short length scales $(<\xi)$. This scaling behavior is satisfied by the simple Lorentzian form [6]

$$C(q) = \frac{1}{2\pi} \frac{w^2 \xi^2}{(1 + a q^2 \xi^2)^{1+H}}, \qquad (6)$$

with $a = (1/2H)[1 - (1 + a Q_c^2 \xi^2)^{-H}]$ if $0 < H < 1$ (power-law roughness), and $a = (1/2)\ln[1 + a Q_c^2 \xi^2]$ if $H = 0$ (logarithmic roughness) [6]. Equation (6) is one possible way of introducing the long-distance cut-off length ξ. We have $Q_c = \pi/a_0$ with a_0 of the order of atomic dimensions, while the parameter w is the rms roughness amplitude. For other self-affine roughness correlation models see also Refs. [4, 7].
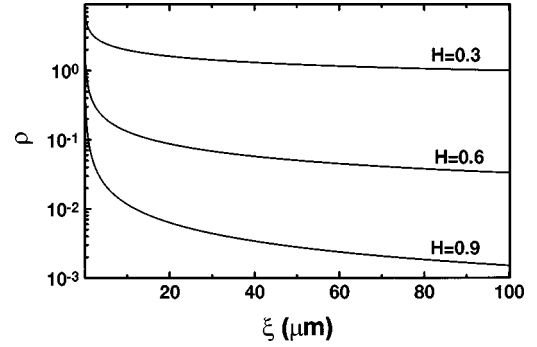

FIG. 2. Local surface slope ρ as a function of the in-plane roughness correlation length ξ for $w = 10$ nm, and various roughness exponents H.

IV. RESULTS AND DISCUSSION

Substitution of Eq. (6) into the expression $\rho \leq \sqrt{\langle (\nabla h)^2 \rangle}$ for the local surface slope yields the simple analytic expression

$$\rho = \frac{w}{\sqrt{2} a \xi} \left[\frac{1}{1-H} \{ T_c^{1-H} - T_L^{1-H} \} + \frac{1}{H} \{ T_c^{-H} - T_L^{-H} \} \right]^{1/2}, \qquad (7)$$

with $T_c = (1 + a Q_c^2 \xi^2)$ and $T_L = (1 + a Q_L^2 \xi^2)$, L is the lateral dimension of $A_{\text{flat}} (\approx L^2)$ and $Q_L = 2\pi/L$. For $H = 0$ and $H = 1$ we obtain from Eq. (7) the limiting forms, if we take the identity $\ln(T) = \lim_{u \to o} (1/u)(T^u - 1)$, then

$$\rho_{H=0} = \frac{w}{\sqrt{2} a \xi} \left[a(Q_c^2 - Q_L^2)\xi^2 + \ln\left(\frac{T_c}{T_L}\right) \right]^{1/2}, \qquad (8)$$

$$\rho_{H=1} = \frac{w}{\sqrt{2} a \xi} \left[\ln\left(\frac{T_c}{T_L}\right) - a(Q_C^2 - Q_L^2)\xi^2 (T_C T_L)^{-1} \right]. \qquad (9)$$

Figure 2 shows the calculations of the local surface slope. Clearly a strong influence of the roughness exponent H is observed.

A. Free energy calculations

The change in the total free energy when the polymer block is in contact with the rough substrate is given by the sum of the adhesive and elastic energy such that [5]

$$U_{ad} + U_{el} = -A_{\text{flat}} \Delta\gamma_{eff},$$

with

$$\Delta\gamma_{eff} = \Delta\gamma \int_0^{+\infty} du (1 + \rho^2 u)^{1/2} e^{-u}$$

$$- \frac{\pi E}{2(1-\nu^2)} \int_{Q_L}^{Q_e} q^2 C(q) dq, \qquad (10)$$

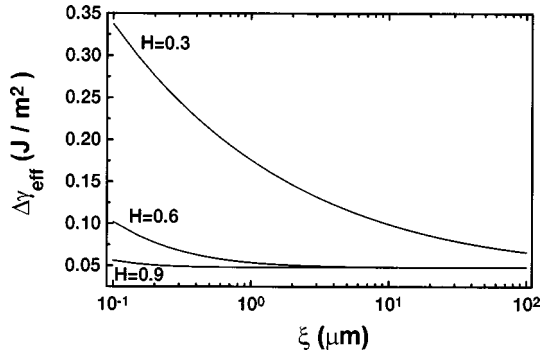where $\Delta\gamma_{eff}$ is the effective change in surface free energy due to substrate surface roughness.

FIG. 3. Effective surface energy $\Delta \gamma_{eff}$ versus in-plane roughness correlation length ξ for various roughness exponents H. $w = 10$ nm, $\Delta \gamma = 4.8 \times 10^{-2}$ J/m^2, $\nu = 0.4$, $E = 1$ MPa, and $L = 1000$ μm ($\gg \xi$).

If the following calculations were performed for $\Delta \gamma = 4.810^{-2}$ J/m^2 [5], rms roughness amplitude $w = 10$ nm and $a_0 = 0.3$ nm. For $\Delta \gamma_{eff}$ the main roughness contribution comes from the local surface slope ρ especially for $E = 0$ (absence of interfacial elastic energy stored in the system), which is the case of polymer adhesives deposited in liquid-like form on solid surfaces followed by drying (although a shrinkage stress may develop which will diminish the adhesion). Moreover, since $C(q) \propto w^2$, the influence of the rms roughness amplitude w on $\Delta \gamma_{eff}$ is rather simple ($\Delta \gamma_{eff} \propto w^2$) for small w (for large w the contribution to adhesion is proportional to w), while any complex dependence on the substrate surface roughness will arise solely from the roughness parameters H and ξ.

Because in Eq. (10) the elastic energy term for $\Delta \gamma_{eff}$ is valid for small local slopes ρ, we show in Fig. 2 the development of the local slope as a function of correlation length ξ for various roughness exponents H. Depending on the value of the rms roughness amplitude w ($\rho \propto w$), the local slope can be significantly large for small roughness exponents setting into question the validity of Eq. (10), except in the case where $E = 0$. The latter reflects the absence of elastic effects, i.e., polymer films formed by the melting of adhesives onto substrates. Therefore, prior to a comparison of theoretical predictions with experimental data (for $E > 0$), an estimation of the local surface slope is highly desirable. The latter requires a knowledge of all three roughness parameters H, w, and ξ as Eq. (7) indicates. Notably, correlation function measurement in terms, i.e., of scanning probe microscopy allows measurement of H, w, and ξ [3,4,6,8,9].

We should point out that the elastic energy contribution in Eq. (10) does not play an important role in the limit of weak roughness or $\rho < 1$ for relatively soft materials ($E < 1$ MPa) which makes the calculations for $\Delta \gamma_{eff}$ relatively reliable even for large local surface slopes ($\rho > 1$; small H and/or large ratios w/ξ), because the adhesive term expression is valid also for strong roughness. As Fig. 3 indicates, the effective free energy is strongly influenced by the surface roughness especially for small roughness exponents ($H < 0.5$) and/or large roughness ratios w/ξ (typically ~ 0.1).

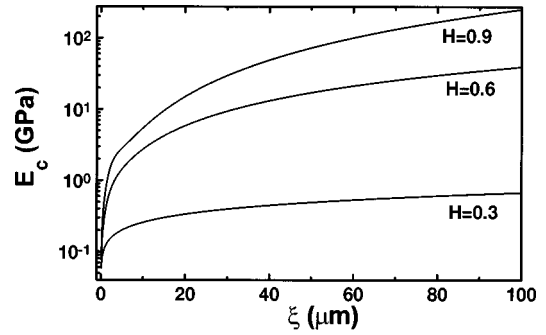The critical elastic modulus E_c for which $\Delta \gamma_{eff} = 0$ is given by

FIG. 4. Critical elastic modulus E_c versus in-plane roughness correlation length ξ for various roughness exponents H. $w = 10$ nm, $\Delta \gamma = 4.8 \times 10^{-2}$ J/m^2, $\nu = 0.4$, and $L = 1000$ μm ($\gg \xi$).

$$E_c = \frac{2(1-\nu^2)\Delta \gamma \int_0^\infty du (1+\rho^2 u)^{1/2} e^{-u}}{\pi \int_{Q_L}^{Q_c} q^2 C(q) dq}, \qquad (11)$$

and varies strongly with variations of substrate roughness in such a way that E_c increases for smoother surfaces, i.e., with an increasing exponent H and/or decreasing roughness ratio w/ξ, see Fig. 4. For elastic modulus $E > E_c$ we have $\Delta \gamma_{eff} < 0$ leading to spontaneous decohesion of the elastic film without an application of any force, whereas for $E < E_c$ a finite force will be necessary to decohere the elastic film. When the elastic energy becomes very high, partial detachments should be considered in order to correctly describe the decohesion process that does not correspond to the vanishing $\Delta \gamma_{eff}$ at a macroscopic scale [Eq. (11)].

B. Force calculations

Assuming a slab of thickness d that undergoes a displacement u upon the action of a force F (Fig. 1). We can calculate the necessary force F to delaminate the film from the substrate by equalizing the elastic energy $A_{flat} d (1/2) E (u/d)^2$ with the effective adhesion energy $A_{flat} \Delta \gamma_{eff}$, which is actually a Griffith calculation in fracture mechanics. Therefore, since $F = A_{flat} E(u/d)$, we obtain

$$F = F_{flat} \left[\int_0^{+\infty} du (1+\rho u)^{1/2} \right.$$

$$\left. \times e^{-u} - \frac{\pi E}{2(1-\nu^2)\Delta \gamma} \int_{Q_L}^{Q_c} q^2 C(q) dq \right]^{1/2}, \qquad (12)$$

with $F_{flat} = A_{flat}(2\Delta \gamma E/d)^{1/2}$. Calculations of the force F for various roughness exponents are depicted in Fig. 5. Clearly for rougher substrates at short length scales a larger force will be required to delaminate the elastic film. Equation (12) is valid for constant strain fields in the polymer film, which is the case for the planar geometry under consideration. In the case of positive moduli and surface energies the presence of roughness leads to an increase in adhesion energy, leading to $F \geq F_{flat}$, where the increment of the adhesion energy is lowered by the elastic term. For other geometries more complex considerations are necessary [5].
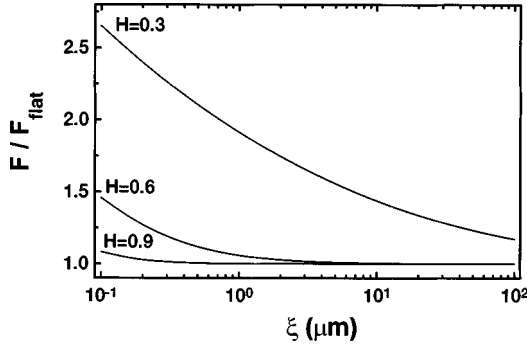
FIG. 5. Effective pull-off force F versus in-plane roughness correlation length ξ for various roughness exponents H. $w=10$ nm, $\Delta\gamma=4.8\times10^{-2}$ J/m^2, $\nu=0.4$, $E=1$ MPa, and $L=1000$ μm ($\gg\xi$).

C. Analytic expressions for adhesive and elastic terms

For small local surface slopes such that $\rho<1$, we can rewrite the integral for the adhesive term [Eq. (10)] in a closed integral form [10]

$$\int_0^{+\infty} du(1+\rho^2 u)^{1/2} e^{-u} = \rho e^{\rho^{-2}}\Gamma(\tfrac{3}{2},\rho^{-2}). \quad (13)$$

$\Gamma(x,a)$ represents the incomplete Γ function. For small ρ the asymptotic expansion leads to

$$\rho e^{\rho^{-2}}\Gamma(\tfrac{3}{2},\rho^{-2}) = 1 + \frac{1}{2}\rho^2 + \sum_{n=2}^{+\infty} R(n)\rho^{2n}, \quad (14)$$

with $R(n)=\{1\times1\times3\times5...(2n-3)\}(-1)^{n-1}/2^n$. However, for the elastic term we have an analytic expression only for roughness exponents $H=0$, $H=0.5$, and $H=1$. If we define the quantities

$$G_1(Q_L,Q_c) = a^{-1/2}\xi^{-1}[\tan^{-1}(\sqrt{a}\xi Q_c) - \tan^{-1}(\sqrt{a}\xi Q_L)],$$

$$G_2(Q_L,Q_c) = (1/2a\xi^2)[Q_c(1+a\xi^2 Q_c^2)^{-1} - Q_L(1+a\xi^2 Q_L^2)^{-1}],$$

$$G_3(Q_L,Q_c) = a^{-1}\xi^{-2}(Q_c-Q_L), \quad (15)$$

$$G_4(Q_L,Q_c) = (1/a\xi^2)[Q_c(1+a\xi^2 Q_c^2)^{-1/2} - Q_L(1+a\xi^2 Q_L^2)^{-1/2}],$$

$$G_5(Q_L,Q_c) = (1/a^{3/2}\xi^3)[\sinh^{-1}(\sqrt{a}\xi Q_c) - \sinh^{-1}(\sqrt{a}\xi Q_L)],$$

we obtain the following expression:

$$G_{el} = \int_{Q_L}^{Q_c} Q^2 C(q)dq = \frac{w^2\xi^2}{2\pi}\begin{cases} G_3(Q_L,Q_c) - a^{-1}\xi^{-2}G_1(Q_L,Q_c) & \text{for} \quad H=0, \\ G_5(Q_L,Q_c) - G_4(Q_L,Q_c) & \text{for} \quad H=0.5, \\ 2^{-1}a^{-1}\xi^{-2}G_1(Q_L,Q_c) - G_2(Q_L,Q_c) & \text{for} \quad H=1. \end{cases} \quad (16)$$

Therefore, from Eqs. (13)–(15) we obtain the simpler expressions

$$\Delta\gamma_{eff} = \Delta\gamma\left\{\rho e^{\rho^{-2}}\Gamma(\tfrac{3}{2},\rho^{-2}) - \frac{\pi E}{2(1-\nu^2)\Delta\gamma}G_{el}\right\}, \quad (17)$$

$$F = F_{flat}\left\{\rho e^{\rho^{-2}}\Gamma(\tfrac{3}{2},\rho^{-2}) - \frac{\pi E}{2(1-\nu^2)\Delta\gamma}G_{el}\right\}^{1/2}. \quad (18)$$

D. Partial contact between layer and substrate

In the previous calculations we assumed a complete contact between the elastic film and the solid substrate. If, however, only partial contact occurs at lateral length scale λ, then the real contact area $A(\lambda)$ is related to the macroscopic nominal contact area $A(L)\cong A_{flat}(\approx L^2, L\gg\xi)$ by the equation

$$A(\lambda) = A_{flat}\frac{2}{\pi}\int_0^{+\infty}\frac{\sin x}{x}e^{-x^2 G(\lambda)}dx = A_{flat}\,\text{erf}\left(\frac{1}{2\sqrt{G(\lambda)}}\right), \quad (19)$$

with

$$G(\lambda) = \frac{\pi}{4}\left[\frac{E}{(1-\nu^2)\sigma_o}\right]^2\int_{2\pi/L}^{2\pi/\lambda} q^3 C(q)dq, \quad (20)$$

with σ_o being the applied load used to press the film onto the hard solid substrate. This represents the situation of a smooth surface on all length scales shorter than λ or an apparent area of contact on the length scale λ Equation (20) assumes conditions of frictionless contact between two elastic solids with rough surfaces. Then, the contact stresses depend only on the shape of the gap between the solids prior to any loading [4]. The stress distribution in the contact area described by the differential equation [11]

$$\frac{\partial P}{\partial\lambda} = \left(\frac{dG}{d\lambda}\right)\sigma_o^2\frac{\partial^2 P}{\partial\sigma^2}. \quad (21)$$

The solution of Eq. (21) with the boundary conditions $P(\sigma=0,\lambda)=0$ (absence of adhesion) and $P(\sigma=+\infty,\lambda)=\infty$

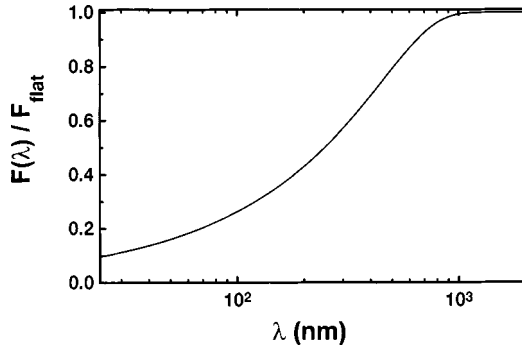FIG. 6. Effective pull-off force $F(\lambda)$ versus contact lateral length scale λ for in-plane roughness correlation length $\xi = 200$ nm, $w = 10$ nm, roughness exponent $H = 0.3$, $\Delta\gamma = 4.8 \times 10^{-2}$ J/m^2, $\nu = 0.3$, $E = 1$ MPa, $E/\sigma_o = 30$, and $L = 1000$ μm $(\gg \xi)$.

yields Eq. (19) with $P(\lambda) = A(\lambda)/A_{\text{flat}}$. If we substitute Eq. (6) for $C(q)$ into Eq. (20) we obtain for $G(\lambda)$ the analytic expression

$$G(\lambda) = \frac{1}{8}\left[\frac{E}{(1-\nu^2)\sigma_o}\right]^2 \rho_\lambda^2, \tag{22}$$

with

$$\rho_\lambda = \frac{w}{\sqrt{2}a\xi}\left[\frac{1}{1-H}\{T_c^{1-H} - T_\lambda^{1-H}\} + \frac{1}{H}\{T_c^{-H} - T_\lambda^{-H}\}\right]^{1/2}, \tag{23}$$

with $T_\lambda = (1 + aQ_\lambda^2\xi^2)$ and $Q_\lambda = 2\pi/\lambda$. Moreover, the effective surface energy $\Delta\gamma_{\text{eff}}$ and the force F to delaminate the film are, respectively,

$$\Delta\gamma_{eff}(\lambda) = \Delta\gamma\rho e^{\rho^{-2}}\Gamma(\tfrac{3}{2}, \rho^{-2}) - \frac{\pi E}{2(1-\nu^2)}\int_{Q_\lambda}^{Q_c} q^2 C(q)dq \tag{24}$$

and

$$F(\lambda) = F_{\text{flat}}\,\text{erf}\left(\frac{1}{2\sqrt{G(\lambda)}}\right)\left[\rho e^{\rho^{-2}}\Gamma(\tfrac{3}{2}, \rho^{-2})\right.$$
$$\left. - \frac{\pi E}{2(1-\nu^2)\Delta\gamma}\int_{Q_\lambda}^{Q_c} q^2 C(q)dq\right]^{1/2}. \tag{25}$$

In this case the effective surface energy $\Delta\gamma_{\text{eff}}$ and the pull-off force will be lower than that of complete contact. Especially for the pull-off force this is depicted in Fig. 6 as a function of contact lateral length scale λ. Clearly with diminishing contact, that is with decreasing λ the force drastically decreases in agreement with predictions of Fuller and Tabor [1]. Notably in Eq. (25) the dominant contribution comes from the reduction of the contact area with decreasing contact length scale λ. The treatment of the partial contact is approximate and a more rigorous treatment is presented in Ref. [14].

V. CONCLUSIONS

It is shown that the surface roughness affects considerably the adhesion forces between an elastic polymer and a rough solid substrate. Analytical calculations of the local surface slope allows an estimation of the roughness effects on the adhesion energy more precisely than those presented in earlier works, especially for roughness exponents $H < 0.5$. For weak surface roughness ($\rho < 1$), the elastic energy contribution is significant on the film effective surface energy $\Delta\gamma_{\text{eff}}$ and pull-off force for elastic modulus E in the range of GPa. Moreover, in the case of partial contact an estimation of the pull-off force shows that it strongly decreases with reducing contact area due to surface. Our calculations are strictly valid for elastic solids, while for in real polymers [12] viscous-elastic effects are present which may alter the value of $\Delta\gamma$. In this case modifications are required since surface roughness introduces fluctuating forces with a wide distribution of frequencies [13].

Finally, we should point out that the calculation of the roughness influence is performed in terms of a specific roughness model [6] which gives an analytical form of the local surface slope by incorporating the effect of intermediate lateral roughness wavelengths $q \sim 2\pi/\xi$, as well as yields analytic results for the elastic contribution for roughness exponents $H = 0$, 0.5, and 1. Clearly for other correlation models [4,7] there can be deviations, because they differ mainly around the lateral roughness wavelengths $q \sim 2\pi/\xi$. However, these effects are lower in significance than errors introduced by the use of only the power-law regime of the roughness spectrum, $C(q) \propto q^{-2-2H}$ [5].

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