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# On stickiness of multiscale randomly rough surfaces

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## Abstract

A new stickiness criterion for solids having random fractal roughness is derived using Persson's theory with DMT-type adhesion. As expected, we find stickiness, i.e., the possibility to sustain macroscopic tensile pressures or else non-zero contact area without load, is not affected by the truncation of the PSD spectrum of roughness at short wavelengths and can persist up to roughness amplitudes orders of magnitude larger than the range of attractive forces. With typical nanometer values of the latter, the criterion gives justification to the well-known empirical Dalhquist criterion for stickiness that demands adhesives to have elastic modulus lower than about 1 MPa.

Keywords: *stickiness criterion, adhesion, Dalhquist criterion, DMT model*

## I. INTRODUCTION

Contact mechanics with roughness has made tremendous progress in recent years (for two recent reviews, see Ref. [1, 2]), and adhesion has become increasingly relevant with the interest on soft materials [3], nano-systems [4, 5] and the analysis of bio-attachments [6, 7]. Contact between solids occurs via large van der Waals forces, usually represented, for example, by the well known Lennard-Jones force-separation law. These forces give rise to a theoretical strength much higher than the typical values to break bulk materials apart. Hence, the "adhesion paradox" [8] states that all objects in the Universe should stick to each other. This does not happen due to inevitable surface roughness at the interface, and Nature has developed different strategies to achieve stickiness, including contact splitting and hierarchical structures [9–11]. At macroscale and for nominally flat bulk solids, it appears that the only solution to maintain stickiness is to reduce the elastic modulus. This is well known in the world of Pressure-Sensitive Adhesives (PSA), soft polymers showing instantaneous adhesion on most surfaces, upon application of just a light pressure [12, 13]. Dahlquist [14, 15] proposed that to achieve a universal stickiness, the elastic Young modulus should be smaller than about 1 MPa (at 1 Hz, as adhesive are strongly viscoelastic their modulus depends on frequency). This criterion has no scientific validation, but appears to be largely used in the world of adhesives.

There have been various attempts to study the problem of elastic contact with roughness and adhesion. Fuller and Tabor (FT, [16]) used the Greenwood and Williamson [17] concept of describing a rough surface with a statistical distribution of identical asperities of radius $R$, together with JKR theory for the sphere contact [18]. FT found that adhesion was easily destroyed with root mean square (RMS) amplitude of roughness $h_{\mathrm{rms}}$ of a few micrometers in spherical rubber bodies against rough hard Perspex surfaces. Their theory depends only on a single dimensionless parameter $\theta_{FT} = h_{\mathrm{rms}}^{3/2}\Delta\gamma/\left(R^{1/2}E^{*}\right)$ where $E^{*}$ is the plane strain elastic modulus, $\Delta\gamma$ is the interface energy. The choice of $R$ seems critical in view of its sensitivity to "resolution" or "magnification" [19], i.e., to the shortest wavelength in the roughness spectrum. In the "fractal limit", i.e., for an infinite number of scales, $R \rightarrow 0$, there would be *no stickiness for any surface*, in the sense that the solution would be identical to that without adhesion, irrespective of the geometrical characteristics, like fractal dimension, or RMS amplitude of roughness. Hence, FT apparent good correlation with the theory despite the many limitations (see [20]), may have been due to a fortuitous choice of $R$ at a relatively coarse scale where measurements were made at that time.

The JKR theory is inappropriate when contact spots become very small, and another theory is more promising in this case, which takes the name of DMT, stemming for the original case of the sphere [21]. Joe, Scaraggi and Barber [22] showed that the JKR approach becomes questionable for contact problems involving fine-scale roughness; in such case a solution based on the full Lennard-Jones traction law is more appropriate. Moreover, Violano and Afferrante [23, 24] showed that DMT-type models are accurate in predicting the effective interfacial binding energy, when compared with calculations including the "exact" Lennard-Jones law. DMT makes possible to solve contact problems with adhesion using results from the adhesionless problem, by assuming that the adhesive stresses do not alter the pressure in the contact area (which therefore remains purely under compression, and remains defined in the same way as "repulsive") nor the gaps outside the contact. The external pressure is therefore the difference of the repulsive and an adhesive pressure $p_{ext} = p_{rep} - p_{ad}$.

Pastewka & Robbins ([25]) presented a criterion for adhesion between randomly rough surfaces after interpreting simulations of adhesive rough contact with fractal roughness, which were obtained on spectra of roughness 3 orders of magnitude in wavelengths (from nano to micrometer scale). They determined the surface "stickiness" based on the slope of the curve repulsive area vs external load (a definition which we shall also adopt here) elaborating a simplified DMT-like model that uses only the asymptotic expression for gaps at the edge of the repulsive contact regions, hence defining an 'adhesive boundary layer' that surrounds the 'repulsive' contact zone. They obtained a criterion for stickiness that depends mainly on small scale features of the rough surface, i.e., on local slopes and curvature, and shows to be in agreement with their numerical calculations.

Recent theories by Ciavarella [26] with its BAM model (Bearing Area Model) and Joe, Thouless and Barber (JTB, [27]), estimate the pressure at pull-off between surfaces does not depend much on local slopes and curvature, and while the former theory is a very simple model, the latter involves a full recursive solution using the Lennard-Jones potential. Similar results are found in Ref. [28], where the effect of surface topograghy on the pull-off force is investigated in the framework of a DMT theory. Also, Refs. [29, 30] using JKR framework, suggest that the contact area reaches a constant value as the magnification is increased and full contact occurs at the short length-scale structures of the surface.

In this paper, we use the Persson and Scaraggi theory (PS theory, [31]) based on the DMT assumption, with some refinements [32], to derive a "stickiness" criterion for very broad spectra, typical of real surfaces which can be expected to have features from millimeter (or more) to nanometer scale.

## II. METHODS

### A. The rough contact model

In DMT theories, the adhesive pressure is computed by convolution of the elementary tension-separation law $\sigma_{ad}(g)$ with the distribution of gaps $P(g)$, obtained by a standard non-adhesive contact solution. In general, the Lennard-Jones force-separation law is usually represented as

$$\sigma_{ad}(g) = \frac{8\Delta\gamma}{3\epsilon}\left[\frac{\epsilon^3}{g^3} - \frac{\epsilon^9}{g^9}\right] \tag{1}$$

where $g$ is the local gap, $\Delta\gamma = \int_\epsilon^\infty \sigma(g)dg$ is the *interface energy* or, by definition, the work done per unit area of interface in separating two bodies from the equilibrium position $g = \epsilon$, at which $\sigma_{ad} = 0$. The maximum tensile traction happens at a separation $g = 3^{1/6}\epsilon$ and is $\sigma_{th} = 16\Delta\gamma/\left(9\sqrt{3}\right)\epsilon$. A possible simplification, which will be adopted in the following derivations, is to use a constant force-law [33]. Considering gaps from the equilibrium point namely $u = g - \epsilon$, imposing the same interface energy $\Delta\gamma$,

$$\begin{aligned} \sigma_{ad}(u) &= \sigma_0, \quad u \leq \epsilon \\ \sigma_{ad}(u) &= 0, \quad u > \epsilon \end{aligned} \tag{2}$$

where $\sigma_0 = 9\sqrt{3}\sigma_{th}/16 \simeq \sigma_{th}$ and $\epsilon$ is the same range of attraction, so obviously $\Delta\gamma = \sigma_0\epsilon$.

Notice that if $E^* = E/\left(1-\nu^2\right)$ is the plane strain elastic modulus ($E$ is the Young's modulus and $\nu$ is the Poisson's ratio), then $l_a = \Delta\gamma/E^*$ defines a characteristic adhesion length which can be used to quantify the relative strength of adhesion. The value of $l_a/\epsilon$ is of the order of the fractional change in bond length needed to change the elastic energy by the binding energy, and $l_a/\epsilon \ll 1$. For example, the typical Lennard-Jones description of an interface has $l_a \simeq 0.05\epsilon$. The theoretical strength in this case, $\sigma_0 = l_a E^*/\epsilon = 0.05E^*$ represents a very high value.

In DMT convolution of the elementary tension-separation law $\sigma_{ad}(u)$ with the distribution of gaps $P(u)$, for the Maugis potential, simplifies therefore to

$$p_{ad} = \sigma_0 \int_0^{\epsilon} du P(u) = \sigma_0 \frac{A_{ad}}{A_{nom}} \tag{3}$$

where $A_{ad}$ is the "adhesive" contact area, i.e. the region where tensile stresses are applied, and $A_{nom}$ is the "nominal" or "apparent" contact area. An elaborate expression for $P(u)$ (for the purely repulsive problem, i.e. in the absence of any adhesion) is obtained in Persson's theory (see Appendix - A eq. (29) and [32, 34]).

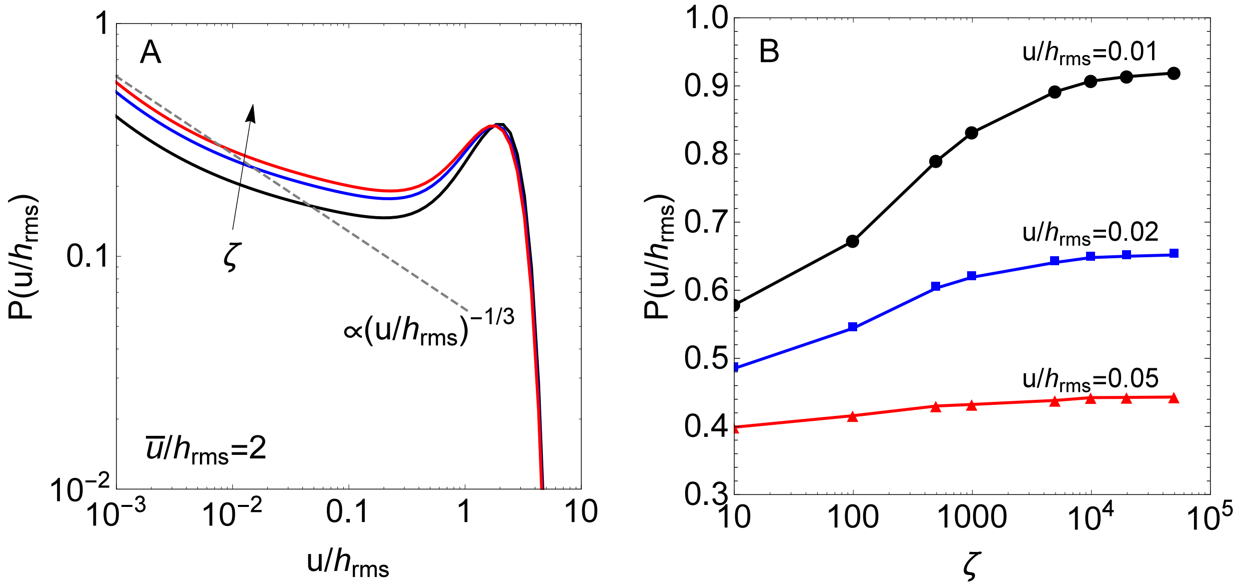


FIG. 1: (A) Distribution of gaps $P(u)$ (black, blue and red line respectively for $\zeta = 10$, 100, 1000) as obtained by Persson's theory. An asymptotic fit $P(u) \sim u^{-1/3}$ is also plotted as guide to the eye (dashed black line). Results are obtained for a self-affine fractal surface with a pure power law PSD, fractal dimension $D = 2.2$ (or Hurst exponent $H = 0.8$), and interfacial mean separation $\overline{u}/h_{rms} = 2$. (B) Dependence of the $P(u)$ on the magnification $\zeta$. Notice a convergence of $P(u)$ with increasing magnifications for various values of the local gap $u/h_{rms}$. Results are obtained for a self-affine fractal surface with a pure power law PSD, fractal dimension $D = 2.2$ (or Hurst exponent $H = 0.8$), and interfacial mean separation $\overline{u}/h_{rms} = 1.37$.

Fig. 1A-B shows that there is a convergence in the distribution of $P(u)$ for increasing magnification $\zeta = q_1/q_0$, being $q_1$ and $q_0$ respectively the high and low wavenumber cut-off of the surface Power Spectral Density (PSD). Furthermore there is an asymptotic scaling at low separations $P(u) \sim u^{-1/3}$. Results in Fig. 1A are given for pure power law PSD, fractal dimension $D = 2.2$ (i.e. Hurst exponent $H = 3 - D = 0.8$) and for a mean gap $\overline{u}$ equal to twice the RMS amplitude of roughness, $\overline{u}/h_{rms} = 2$. We define a non-dimensional pressure $\widehat{p}_{rep} = p_{rep}/\left(E^* q_0 h_{rms}\right)$ and we remark that, for typical real surfaces $H \gtrsim 0.6$, in the limit of relatively large $\zeta$ and small pressures, Persson's theory reduces to $\widehat{p}_{rep} \simeq \frac{3}{4}\exp\left(-2\overline{u}/h_{rms}\right)$ [35, 36]. As we are essentially interested in the region of the area-load relationship near the axes origin, we disregard $\overline{u}/h_{rms} < 1$, and also $\overline{u}/h_{rms} > 3$ where we are likely to have finite effects due to poor statistics of the Gaussian surfaces and very few asperities in contact. This corresponds therefore to the range $\widehat{p}_{rep} = 10^{-3} - 10^{-1}$.

Furthermore, within our DMT hypothesis, the range of attractive forces of interest is $\epsilon << h_{rms}$, where we can assume that the main contribution to the gaps and hence to adhesion comes from the asymptotic value of $P(u)$ at low $u$, namely from the regions close to the contact boundaries. We can obtain more understanding of this asymptotic form of $P(u)$ from standard contact mechanics theory, and from the asymptotic part of the original Persson's theory [37], whereas we shall use the full Persson and Scaraggi theory only for the actual calculation of the prefactors. Notice that these asymptotic derivations are similar to what suggested by Pastewka and Robbins [25], except that we shall not make their assumptions, which lead to the different final results.

### B. Asymptotic results

Here we derive an asymptotic theory for the attractive area $A_{ad}$. It is well known that the relationship between (repulsive) contact area ratio $A_{rep}/A_{nom}$ and mean pressure $p_{rep}$

$$\frac{A_{rep}}{A_{nom}} \simeq \kappa_{rep} \frac{p_{rep}}{E^* \sqrt{2m_2}} \tag{4}$$

is linear for $A_{rep}/A_{nom}$ up to almost $20\% - 30\%$ [37]. In the original Persson's theory, $\kappa_{rep} = \frac{2\sqrt{2}}{\sqrt{\pi}} = 1.6$, and we can also write $V = \frac{1}{2}E^{*2}m_2$ as the variance of full contact pressures. Here, $m_2$ is the mean square profile slope along any direction (for a isotropic surface). The distribution of pressures $P(p)$ near the boundaries of contact (on the contact side) is at low $p$ [38]

$$P(p) \simeq \frac{p\, p_{rep}}{V} \sqrt{\frac{2}{\pi V}} \tag{5}$$

Suppose the perimeter of the actual contact area [not necessarily simply-connected] is $\Pi$.

We define position on $\Pi$ by a curvilinear coordinate $s$. In view of the asymptotic behaviour at the edge of the contact area [39], we must have at every point on $\Pi$, pressure $p$ and gap $u$ as

$$p(x) = B(s)\, x^{1/2}; \qquad u(x) = C(s)\, x^{3/2}; \tag{6}$$

where $x$ is a coordinate perpendicular to the boundary and $C(s) = \beta d(s)^{-1/2}$ where $\beta$ is a geometrical prefactor of order 1, $B(s) = 3E^* \beta d(s)^{-1/2}/4$ and $d(s)$ is a local characteristic length scale.

It is clear that, as load is increased, existing contacts grow larger and new contacts form, resulting in a very irregular shape. Already Greenwood and Williamson [17] in their simple asperity theory suggested that the *average* radius of contact should remain constant with load, as a result of competition between growing contacts and new contacts forming, and this is correctly captured despite the strong approximations in the asperity model. In Ref. [40] it was shown, again with a simple asperity model, but with an exponential distribution of asperity heights, $d$ seems indeed completely independent on load.

Hence, we shall leave the quantity $d(s)$ to vary arbitrarily along the perimeter. For a segment $ds$ of $\Pi$, there exists a region $dxds = \frac{2pdpds}{B(s)^2}$ in which the pressure is in the range ($p$, $p+dp$). It follows that the total area with the pressure is $\int_S \frac{2pdpds}{B(s)^2}$, and hence the probability of an arbitrary point being in this range is $P(p)\, dp$ and the PDF is

$$P(p) = \frac{2p}{A_{nom}} \int_{\Pi} \frac{ds}{B(s)^2} = \frac{2p\Pi}{A_{nom}} I_p \tag{7}$$

where $I_p = \int_0^1 d\widehat{s}/B(\widehat{s})^2 = [4/(3\beta E^*)]^2 \langle d \rangle$, $\widehat{s} = s/\Pi$ is a normalized coordinate along the perimeter, and $\langle d \rangle$ means the mean value of $d$. A similar argument with the gap expression yields

$$P(u) = \frac{1}{A_{nom}} \left( \frac{4}{9u} \right)^{1/3} \Pi I_u \tag{8}$$

where $I_u = \beta^{-2/3} \int_0^1 d(\widehat{s})^{1/3} d\widehat{s} = \beta^{-2/3} \langle d^{1/3} \rangle$. Eliminating the perimeter from eqs. (7, 8) and using eq. (5), we obtain $\Pi = \frac{pA_{nom}}{2I_p} \sqrt{\frac{2}{\pi V^{3/2}}}$ and hence

$$P(u) = \left( \frac{4}{9u} \right)^{1/3} \frac{I_u}{2I_p} \sqrt{\frac{2}{\pi V^3}} p_{rep} \tag{9}$$

Upon integration of the distribution of gaps $P(u)$, the attractive area can be given as

$$\frac{A_{ad}}{A_{nom}} = \frac{3}{2} a_V \widehat{p}_{rep} \left( \frac{\epsilon}{h_{rms}} \right)^{2/3} \tag{10}$$

being the coefficient $a_V$

$$a_V = \frac{3}{2} \left( \frac{4}{9} \right)^{1/3} \frac{9}{32 \beta^{8/3}} \frac{\left\langle d^{1/3} \right\rangle}{\langle d \rangle} E^{*3} \sqrt{\frac{2}{\pi V^3}} q_0 h_{rms}^{5/3} \tag{11}$$

Notice, in eq. (10), $A_{ad}/A_{nom}$ is proportional to the external mean pressure if $\left\langle d^{1/3} \right\rangle / \langle d \rangle$ does not depend on pressure.

## III. RESULTS

We have obtained an expression for $a_V$ in eq. (10), but this was mainly for qualitative purposes because of the problematic term $\left\langle d^{1/3} \right\rangle / \langle d \rangle$. Hence, we fit the global $A_{ad}/A_{nom}$ vs. $(\epsilon/h_{rms})^{2/3}$ curves obtained from numerical results of the adhesionless contact problem with the full Persson-Scaraggi's theory. The use of the Persson-Scaraggi's theory allows us to reach broad band of roughness, $\zeta \simeq 10^5$. Fig. 2 shows that in terms of actual adhesive area, the convergence with $\zeta$ is very rapid (Fig. 2A) and is not modified by the load (Fig. 2B). Accordingly, interpreting the results in terms of the prefactor $a_V$ , the solution rapidly converges with magnification (Fig. 2C), and weakly depends on pressure at the smaller values of $\hat{p}_{rep}$ (Fig. 2D) or indeed on fractal dimension in the range $D = 2.1 - 2.3$, which is the most interesting range [41]. We also show Pastewka and Robbins' prediction for the prefactor $a_V$, which corresponds to $a_{V,PR} \sim \zeta^{1/3}$ for $D = 2.2$ (see Appendix B) and hence may result in large error for large magnifications.

Starting again from eq. (10), we find that both repulsive mean pressure and adhesive mean pressure are proportional to the repulsive contact area, which can therefore be grouped as

$$\begin{aligned} \frac{p_{ext}}{E^*} &= \frac{p_{rep}}{E^*} - \frac{\sigma_0}{E^*} \frac{A_{ad}}{A_{nom}} \\ &= \frac{A_{rep}}{A_{nom}} \frac{\sqrt{2m_2}}{2} \left[ 1 - \frac{l_a}{\epsilon} \frac{3}{2} \frac{a_V}{q_0 h_{rms}} \left( \frac{\epsilon}{h_{rms}} \right)^{2/3} \right] \end{aligned} \tag{12}$$

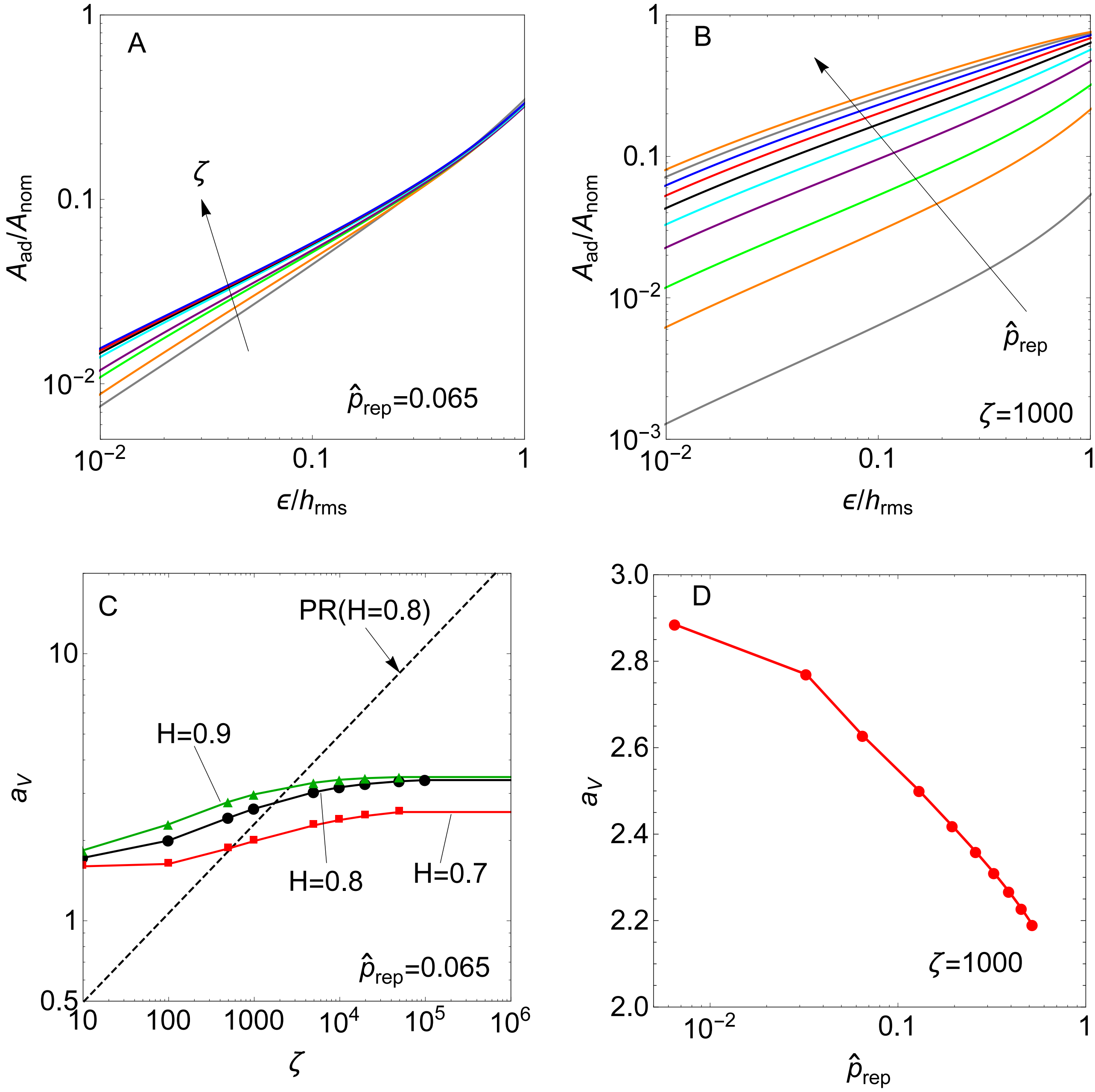


FIG. 2: (A) Adhesive area $A_{ad}/A_{nom}$ estimated by Persson's theory (with $\zeta = [10, 100, 500, 1000, 5000, 10000, 20000, 50000]$ , $H = 0.8$) . (B) Adhesive area $A_{ad}/A_{nom}$ estimated by Persson's theory (with $p_{rep} = [6.5 * 10^{-3}, 0.03, 0.07, 0.13, 0.20, 0.26, 0.33, 0.39, 0.46, 0.52]$ and $\zeta = 1000$). (C) Estimates of the prefactor $a_V$ from Persson's theory for $H = [0.7 - 0.8 - 0.9]$ vs Pastewka and Robbins' estimate for $H = 0.8$ (black dashed line), $[a_V]_{PR} = 0.252\zeta^{1/3}$ as obtained in Appendix - B as a function of magnification $\zeta$ for $\widehat{p}_{rep} = 0.065$. (D) Estimates of the prefactor $a_V$ for $\zeta = 1000$ from Persson's theory as a function of for $\widehat{p}_{rep}$.

where we used the identity $l_a/\epsilon = \sigma_0/E^*$ and the actual value of the factor $\kappa_{rep} = 2$ [42, 43] instead of the factor of the original Persson's theory.

Hence for the "slope" $\kappa$ we write

$$\frac{1}{\kappa} = \frac{1}{\kappa_{rep}} - \frac{1}{\kappa_{ad}} = \frac{1}{2} - \frac{3a_V}{4q_0 h_{rms}} \frac{l_a}{\epsilon} \left(\frac{\epsilon}{h_{rms}}\right)^{2/3} \tag{13}$$

and then stickiness is obtained when $1/\kappa < 0$ leading to the suggested criterion

$$\frac{\epsilon}{l_a} \left(\frac{h_{rms}}{\epsilon}\right)^{2/3} < \frac{3}{2} \frac{a_V}{q_0 h_{rms}} \tag{14}$$

In particular, neglecting the weak dependence on fractal dimension (see Fig. 2C), considering we are interested in the asymptotic regime of low pressure (see Fig. 2D) and in the limit of high magnification (see Fig. 2C), we can take $a_V \simeq 3$ and rewrite the criterion given in eq. (14) as

$$\frac{h_{rms}}{\epsilon} < \left(\frac{9}{4} \frac{l_a/\epsilon}{\epsilon q_0}\right)^{3/5} \tag{15}$$

As it can be seen eq. (15) depends *only* on RMS amplitude of roughness $h_{rms}$ and the largest wavelength in the roughness spectrum $q_0$.

The obtained criterion depends only on quantitative macroscopic entities, rather than local, small-wavelength dependent ones. The small wavevector cutoff of roughness $q_0$, could in principle take arbitrarily low values for large surfaces, provided that for a given $h_{rms}/\epsilon$ the PSD components should be lowered over all the wavenumber $q$, which would result in increasingly loose boundary of stickiness. For example, for a flat surface of mm size, we have $\epsilon q_0 \sim 10^{-6}$ and our criterion gives $h_{rms}/\epsilon \lesssim 1000$, so macroscopic stickiness could persist even for roughness three orders of magnitude larger than the range of attraction, i.e. of the order of nearly one micron. Of course these extrapolations will have some limitation on the concept of the ideally flat surface with a pure power law PSD of roughness.

Sticky surfaces in Pastewka and Robbins's simulations [25] have $l_a/\epsilon = 0.05$, $\epsilon q_0 = 2\pi/4096$, hence the right hand side of eq. (15) gives $\left(\frac{9}{4}\frac{l_a/\epsilon}{\epsilon q_0}\right)^{3/5} \simeq 13$, while from [44, 45] it is possible to estimate $h_{rms}/\epsilon \simeq [5 - 10]$ which means our stickiness criterion (eq. (15)) was also satisfied for their "sticky surfaces". Nevertheless the criterion we propose does not depend on small scale features of the rough surface and coincides with that of Pastewka &

Robbins [25] only for $\zeta \approx 10^3$ (see Fig. 2C and for a more detailed comparison see Appendix - B).

Obviously, our criterion may suffer from the limitations and assumptions of Persson and Scaraggi's theory, in particular those of the DMT theory. Indeed, from JTB's results [27], we know that complex instabilities and patterns form at very low RMS amplitude of roughness, and in particular that any DMT type of analysis, including the one presented here, can be expected to hold only for approximately

$$\frac{h_{rms}}{\epsilon} > \frac{4}{75}\frac{l_a/\epsilon}{\epsilon q_0} \tag{16}$$

It is also likely, from JTB predictions [27], that stickiness is lower if this condition is violated and a more refined analysis is needed, which is outside the possibilities of both JTB and our model. Fig. 3 shows the stickiness map obtained with the present criterion. The shaded region in Fig. 3A identifies the couple $(\frac{\epsilon q_0}{l_a/\epsilon}, \frac{h_{rms}}{\epsilon})$ which would give stickiness. Underneath the dashed line (from eq. (16)) pattern formation is expected, and our DMT model may not be accurate. In Fig. 3B the slope angle $\alpha = \arctan(\kappa)$ is plotted as a function of $h_{rms}/\epsilon$ for varying $\frac{\epsilon q_0}{l_a/\epsilon}$.

## IV. DISCUSSION

Considering our result, it is clear that to improve stickiness, for a given range of attractive forces $\epsilon$, we need to make $q_0$ as small as possible for the given $h_{rms}$. For a power law PSD $C(q) = C_0 q^{-2(H+1)}$, we have $h_{rms} \simeq \sqrt{\pi C_0/H} q_0^{-H}$. It may be useful to rewrite the criterion (15) in terms of the PSD multiplier $C_0$ (as usual, for $H = 0.8$)

$$C_0 < \frac{0.8}{\pi}\epsilon^{4/5} q_0^{2/5} \left(\frac{9}{4}\frac{l_a}{\epsilon}\right)^{6/5} \tag{17}$$

It appears clear that we need as small roughness as possible, for a given $q_0$ which is presumably dictated by size of the specimen up to some extent, or by the process from which the surface originates. Also, we need to have $l_a/\epsilon$ as high as possible, and this means obviously high $\Delta\gamma$ and low $E^*$ (being $l_a = \Delta\gamma/E^*$).

Given $\Delta\gamma$ is in practice strongly reduced by contaminants and various other effects to

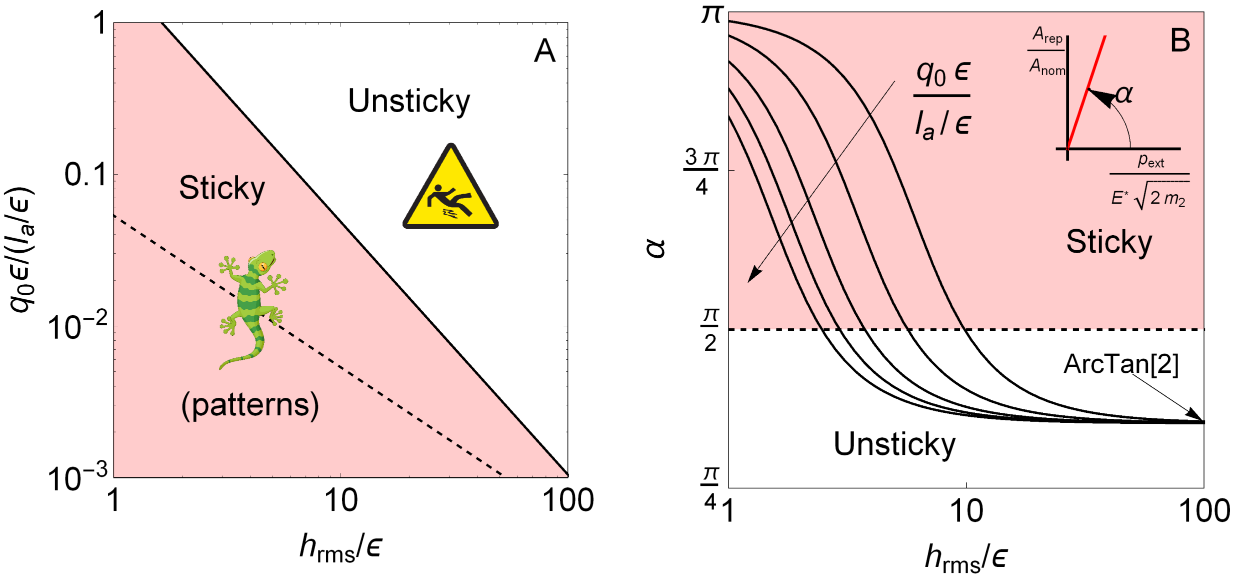


FIG. 3: (A) Adhesion map for multiscale rough surfaces according to the present criterion (15). In the plane $\frac{\epsilon q_0}{l_a/\epsilon}$ vs $\frac{h_{rms}}{\epsilon}$ the sticky region is shaded. According to JTB analysis in the region below the dashed black line pattern formation is expected with possible reduction of stickiness. (B) The slope angle $\alpha = \arctan(\kappa)$ is plotted as a function of $h_{rms}/\epsilon$ for varying $\frac{\epsilon q_0}{l_a/\epsilon} = [1, 3/4, 1/2, 1/4, 1/10]$ and $a_V = 3$. The arrow denotes increasing values of $\frac{\epsilon q_0}{l_a/\epsilon}$.

values of the order $\sim 50$ mJ/m$^2$, the only reliable way to have high stickiness is to have very soft materials.

Specifically, we shall make some quantitative estimates based on real surfaces. As reported by Persson [41], most polished steel surfaces for example, when measured on $L \sim 0.1$ mm, show $h_{rms} \sim 1$ $\mu$m. This means $q_0 h_{rms} \sim 0.1$. This incidentally satisfies JTB condition for "DMT"-behaviour (eq. (16)). Contrary to the recent emphasys on measuring entire PSD of surfaces, it seems therefore that for stickiness, the most important factors are well defined macroscopic quantities, which are easy to measure. For a wide range of surfaces (asphalt, sandblasted PMMA, polished steel, tape, glass) reported in [41], see Fig. 4, $C_0$ is at most $5 \times 10^5$ m$^{0.4}$. Then, assuming $\epsilon = 0.2$ nm and $\Delta\gamma = 50$ mJ/m$^2$ as a typical value, even with the uncertainty in the choice of $q_0$, in the range of $q_0 \sim 10^3$ [m$^{-1}$], our criterion predicts $E^* < 2.4$ MPa. This is for asphalt which clearly is one of the most rough surface we can consider, and indeed outside the normal application of PSA. Our result is therefore entirely compatible with the empirical criterion by Carl Dahlquist [14] from 3M which suggests to make tapes only with low modulus materials $E^* < 1$ MPa, whose generality was so far still scientifically unexplained.

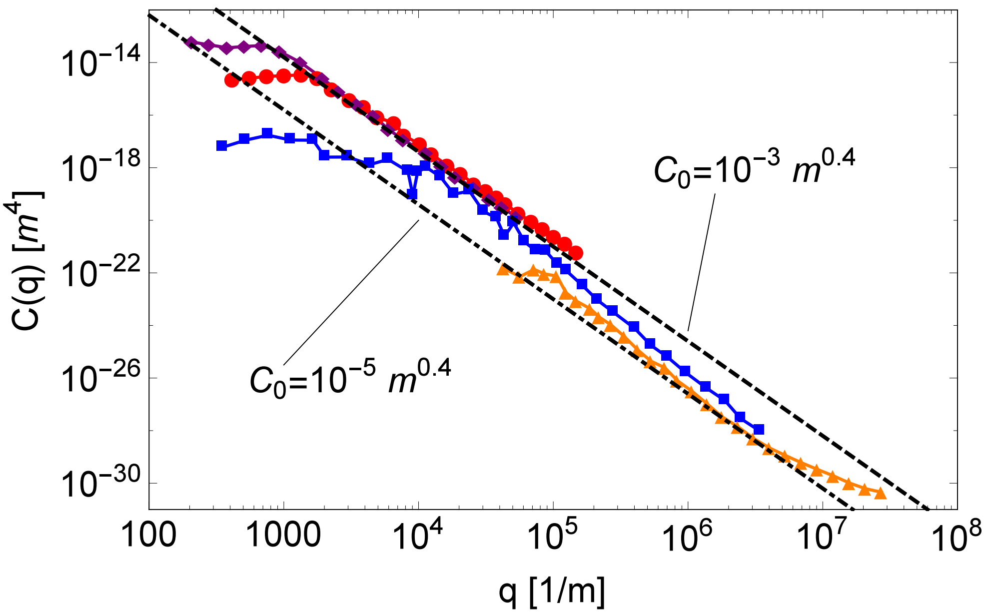


FIG. 4: Experimentally measured PSD of typical real surfaces as taken from Ref. [41]. Notice they can be grouped in a fairly narrow band.

Recently, after a recent paper by Dalvi et al. [48], which has demonstrated that the simple energy idea of Persson and Tosatti [49] works reasonably well, Ciavarella [50] compared the present DMT criterion with other ideas, namely one coming from an energy balance due to Persson and Tosatti [49], where the reduction in apparent work of adhesion equals the energy required to achieve conformal contact, and another criterion derived from BAM (Bearing Area Model) of Ref. [26]. It was found that the three criteria give very close results but the present criterion contains a slightly different qualitative dependence on material properties than others, since we can write it in the form

$$h_{rms} < \epsilon^{-1/5} \left(0.36 l_a \lambda_L\right)^{3/5} \tag{18}$$

being $\lambda_L = 2\pi/q_0$. The above formula shows the product $l_a \lambda_L$, is raised to the power 3/5, and there is a weak apparent dependence on the range of attractive forces $\epsilon$.

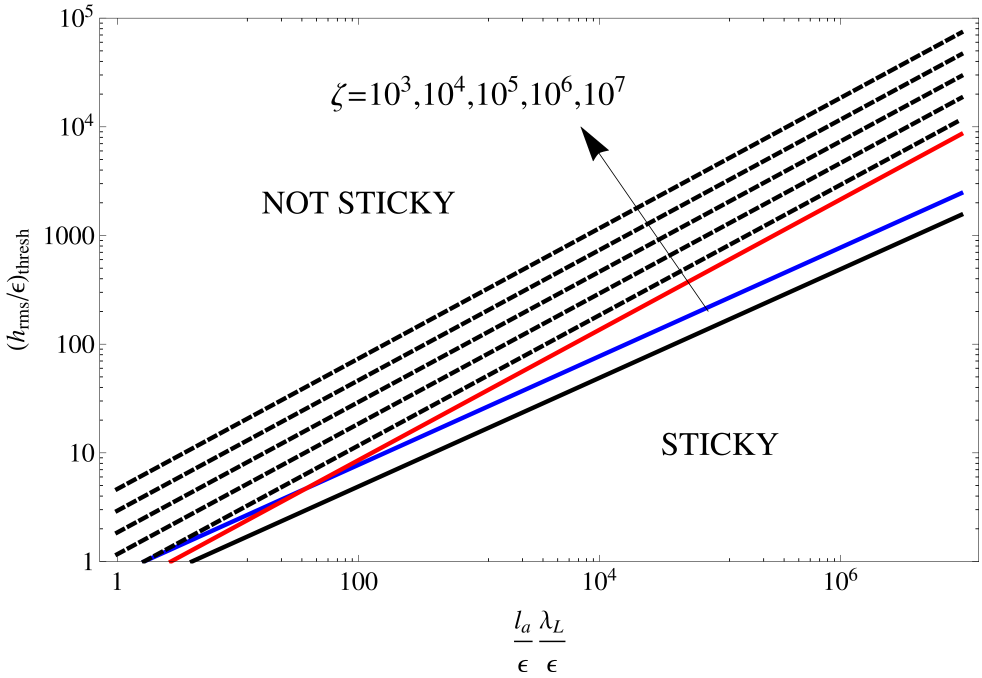


FIG. 5: A comparison of the derived stickiness criterion (red line), together with Persson-Tosatti (black line), BAM (blue solid line) and that of Pastewka and Robbins [25] (black dashed lines) which correlates well with ours only for $\zeta < 1000$.

We can summarize the three criteria for $H = 0.8$ in the form

$$\frac{h_{rms}}{\epsilon} < \sqrt{0.24 \frac{l_a}{\epsilon} \frac{\lambda_L}{\epsilon}} \quad ; \quad \text{Persson-Tosatti} \tag{19}$$

$$\frac{h_{rms}}{\epsilon} < \sqrt{0.6 \frac{l_a}{\epsilon} \frac{\lambda_L}{\epsilon}} \quad ; \quad \text{BAM} \tag{20}$$

$$\frac{h_{rms}}{\epsilon} < \left(0.358 \frac{l_a}{\epsilon} \frac{\lambda_L}{\epsilon}\right)^{3/5} \quad ; \quad \text{Violano et al.} \tag{21}$$

and a comparison is shown in Fig. 5, where Persson-Tosatti is reported in black solid line, BAM as blue solid line, and Violano et al. as red solid line.

Clearly, the criterion obtained in the present paper deviates from the other two at high $l_a/\epsilon$, which corresponds to low elastic modulus, where indeed one expects that the DMT assumptions may be violated. Notice that the PR criterion for $H = 0.8$ writes as

$$\frac{h_{rms}}{\epsilon} < \left(0.06 \frac{l_a}{\epsilon} \frac{\lambda_L}{\epsilon}\right)^{3/5} \zeta^{1/5} \tag{22}$$

It is reported in Fig. 5 and shows similar behaviour from ours but only for low magnifications

## V. CONCLUSIONS

We have defined a new stickiness criterion, whose main factors are the low wavevector cutoff of roughness, $q_0$, the RMS amplitude of roughness $h_{rms}$ and the ratio between the work of adhesion and the plain strain Young modulus. We find that, in principle, it is possible to have effective stickiness even with *quite large RMS amplitudes*, orders of magnitude larger than the range of attractive forces. For robust adhesion with different possible levels of roughness, the main characteristic affecting stickiness is the elastic modulus, in qualitative and quantitative agreement with Dahlquist criterion, well-known in the world of pressure-sensitive adhesives. The proposed criterion is independent on small scale features of the roughness, i.e. slopes and curvatures. According to our analysis stickiness may still depend on the latter, but only for narrow PSD spectra. The result provides new insights on a very debated question in the scientific community, and may serve as benchmark for the future analysis with broad PSD spectra that are still beyond the present computational capabilities.

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## Appendix - A

### A brief outline of the Persson's theory

We summarize the main results of Persson's contact mechanics theory, as applied to adhesive problem, in the variant suggested by Ref. [32]. The repulsive contact area relative to the nominal contact area $A_{rep}(\zeta)/A_{nom}$ is related to the mean pressure $p_{rep}$ (original Persson's theory, [37]) as

$$\frac{A_{rep}(\zeta)}{A_{nom}} = erf\left(\frac{p_{rep}}{E^{*}\sqrt{m_{2}(\zeta)}}\right) \tag{23}$$

where $E^*$ is the elastic modulus in plane strain, and $m_2(\zeta)$ variance of profile slopes at the magnification $\zeta = q_1/q_0$, being $q_1$ and $q_0$ respectively the high and low wavenumber cut-off of the surface Power Spectral Density (PSD). A better estimation of the contact area can be obtained (see Ref. [46]) replacing in eq. (23) $m_2$ with $\langle \nabla h^2 \rangle /2$, where for a given PSD spectrum $C(\mathbf{q})$

$$\left\langle \nabla h^2 \right\rangle = \int_{D(\zeta)} d^2q \; q^2 C(\mathbf{q}) \, S(q) \, , \tag{24}$$

being $D(\zeta) = \{\mathbf{q} \in \mathbb{R}^2 | \; q_0 \leq |\mathbf{q}| \leq \zeta q_0\}$, which can be interpreted as the averaged square slope of the deformed surface (with heights $h$), and $S = \gamma + (1-\gamma)(A/A_0)^2$, being $\gamma$ an empirical parameter which can be taken in the range $0.4 - 0.5$.

For small nominal squeezing pressure or, equivalently, very large magnifications, eq. (23) reduces to eq. (4).

At the magnification $\zeta$, in the apparent contact area $A_{rep}(\zeta)/A_{nom}$, the root mean square roughness amplitude of the rough surface is

$$h_{rms}^2(\zeta) = \int_{q>q_0\zeta} d^2qC(q) = 2\pi \int_{q>q_0\zeta} dqqC(q) \, . \tag{25}$$

The corresponding mean interfacial separation $\overline{u}(\zeta)$ between the surfaces is related to the pressure $\overline{p}_{rep}$ by (Ref. [47])

$$\begin{aligned} \bar{u}(\zeta) = & \frac{1}{2\sqrt{\pi}} \int_{D(\zeta)} d^2qqC(\mathbf{q}) \, w(q) \int_{\overline{p}_{rep}}^{\infty} \frac{dp}{p} \\ & \times \left[ \gamma + 3(1-\gamma) \, \mathrm{erf}^2 \left( \frac{w(q) \, p}{E^*} \right) \right] e^{-\left(\frac{w(q)p}{E^*}\right)^2} \end{aligned} \tag{26}$$

where

$$w(q) = \left( \frac{1}{2} \int_{D_q} d^2q' q'^2 C(\mathbf{q}') \right)^{-1/2} \tag{27}$$

being $D_q = \{\mathbf{q} \in \mathbb{R}^{\mathbf{2}} | \; q_0 \leq |\mathbf{q}| \leq q\}$.

In the limit of vanishing pressure, one can show that eq. (26) simplifies as (Ref. [47])

$$p_{rep} \approx \beta(\zeta) \, E^* \exp\left( -\frac{\bar{u}}{u_0(\zeta)} \right) \tag{28}$$

where $u_0(\zeta)$ is a characteristic length which is independent of the squeezing pressure and depends on the surface roughness (and, hence, on the magnification $\zeta$).

The distribution of interfacial separations also depends on magnification, and is obtained by a much more complex process of integration

$$P(u) \simeq \frac{1}{A_0} \int \frac{d\zeta \left[-A'(\zeta)\right]}{\sqrt{2\pi h_{\rm rms}^2(\zeta)}} \left[\exp\left(-\frac{(u-u_1(\zeta))^2}{2h_{\rm rms}^2(\zeta)}\right) + \exp\left(-\frac{(u+u_1(\zeta))^2}{2h_{\rm rms}^2(\zeta)}\right)\right] \tag{29}$$

where the apex symbol denotes differentiation by magnification.

Predictions of the above expression can be improved by substituting $h_{\rm rms}(\zeta)$ with (see Ref. [32])

$$h_{\rm rms}^{\rm eff}(\zeta) = \left[h_{\rm rms}^{-2}(\zeta) + u_1^{-2}(\zeta)\right]^{-1/2} \tag{30}$$

Finally, $u_1(\zeta)$ is the (average) height separating the surfaces which appear to come into contact when the magnification decreases from $\zeta$ to $\zeta - \Delta\zeta$, where $\Delta\zeta$ is a small (infinitesimal) change in the magnification, and can be calculated from the average interfacial separation $\overline{u}(\zeta)$ between the surfaces *in the (apparent) contact regions observed at the magnification* $\zeta$, and the contact area $A(\zeta)$ as

$$u_1(\zeta) = \overline{u}(\zeta) + \overline{u}'(\zeta)\, A'(\zeta)/A(\zeta) \tag{31}$$

**Appendix - B**

**Comparison with Pastewka and Robbins' theory**

The Pastewka and Robbins (PR) criterion [25] is summarized in the following equation (eqt. 10 in their paper)

$$\frac{h'_{\rm rms}\Delta r}{2l_a}\left(\frac{h'_{\rm rms} d_{rep}}{4\Delta r}\right)^{2/3} < \pi\left(\frac{3}{16}\right)^{2/3} \simeq 1 \tag{32}$$

where $d_{rep}$ is a characteristic diameter of repulsive contact areas, which they estimate as $d_{rep} = 4h'_{rms}/h''_{rms}$, and $h'_{rms}$ and $h''_{rms}$ are the RMS slopes and curvature. Finally, $\Delta r$ is the attractive range which is of the order of atomic spacing ($\Delta r \simeq \epsilon$ for the Lennard-Jones potential). When the condition given by eq. (32) is satisfied, the surfaces in mutual contact are suggested to be "sticky" and a finite value of the pull-off force should occur.

However, comparison with the PR results is best done considering the attractive area (their eq. 6), reading

$$A_{ad} = A_{rep}\left(\frac{16}{9\pi}\right)^{-1/3}\left(\frac{\pi\Delta r}{h'_{rms}d_{rep}}\right)^{2/3} \tag{33}$$

$$= 2\left(\frac{16}{9\pi}\right)^{-1/3} p\frac{A_{nom}}{h'_{rms}E^*}\left(\frac{\pi a_0\sqrt{24l_a/\epsilon}\,h''_{rms}}{4h'^2_{rms}}\right)^{2/3} \tag{34}$$

where we used their equation $A_{rep} = 2pA_{nom}/\left(h'_{rms}E^*\right)$ and $\Delta r/\epsilon = \sqrt{24l_a/\epsilon}$ from Supplementary Information of PR paper so that for $l_a/\epsilon = 0.05$. Hence, it can be shown that, compared to our eq. (10), their estimate corresponds to

$$\left[a_V(\zeta)\right]_{PR} = 1.4622q_0h_{rms}\left(\frac{h''^2_{rms}}{h'^7_{rms}}\right)^{1/3} h^{2/3}_{rms} \tag{35}$$

Now for power law tail of the PSD $C(q) = C_0q^{-2(H+1)}$, estimating $h'_{rms} = \sqrt{2m_2}, h''_{rms} = \sqrt{8m_4/3}$ (see Appendix - C) we obtain as for $H = 0.8$

$$\left[a_V(\zeta)\right]_{PR} = 0.252\zeta^{1/3} \tag{36}$$

which results in a magnification dependence much stronger than the dependence we find (see Fig. 2C). Hence, we conclude PR criterion and the one proposed here agree for a narrow range of $\zeta$ ($\simeq 10^3$), but, at very high $\zeta$ ($\simeq 10^6$, see [48]), the difference grow since in our theory the adhesive contact area $A_{ad}/A_{nom}$ converges with $\zeta$ (eq. 10) while PR estimate continues to grow (eq. 36).

## Appendix - C

### On random process theory

Assume the surface $h(x,y)$ has a continuous noise spectrum in two dimensions and is described by a Gaussian stationary process. In such case, we write

$$h(x,y) = \sum_n C_n \cos\left[q_{x,n}x + q_{y,n}y + \phi_n\right] \tag{37}$$

where the wave-components $q_{x,n}$ and $q_{y,n}$ are supposed densely distributed throughout the $(qx, qy)$ plane. The random phases $\phi_n$ are uniformoly distributed in the interval $[0,\ 2\pi)$. The amplitudes $C_n$ are also random variables such that in any element $dq_x dq_y$

$$\sum_n \frac{1}{2} C_n^2 = C(q_x, q_y)\, dq_x dq_y. \tag{38}$$

The function $C(q_x, q_y)$ is the Power Spectral Density (PSD) of the surface $h$, whose mean-square value can been calculated as

$$m_{00} = \int\int_{-\infty}^{+\infty} C(q_x, q_y)\, dq_x dq_y \tag{39}$$

For isotropic roughness, using Nayak [52] definitions for the surface

$$m_{rs} = \int\int_{-\infty}^{\infty} C[q_x, q_y]\, q_x^r q_y^s dq_x dq_y \tag{40}$$

where $m_{00}$ is by definition $h_{rms}^2$. It can be shown by defining the PSD and the ACF (auto-correlation function) of the partial derivatives of $h$ with respect to $x$ and $y$ coordinates, and using a relationship with the PSD of the surface, that the above spectral moments are (see [51])

$$\begin{aligned} \left\langle \left( \frac{\partial^{r+s} h}{\partial x^r \partial y^s} \right)^2 \right\rangle &= m_{2r,2s} \\ \left\langle \left( \frac{\partial^{r+s} h}{\partial x^r \partial y^s} \right)^2 \right\rangle &= (-1)^{\frac{1}{2}(r+s-r'-s')}\, m_{r+r',s+s'} \quad \text{or} \quad 0, \end{aligned} \tag{41}$$

depending on $(s + r - r' - s')$ is even or odd.

Nayak [52] finds for isotropic surface,

$$
\begin{aligned}
& m_{20} = m_{02} = m_2; \qquad m_{11} = m_{13} = m_{31} = 0 \\
& m_{00} = m_0; \qquad 3m_{22} = m_{40} = m_{04} = m_4
\end{aligned}
\tag{42}
$$

meaning when there is no second subscript the *profile statistics for isotropic surface*, which is independent on the direction chosen.

For slopes, with the common definition of their RMS value is (also used by PR [25])

$$
h'_{rms} = \sqrt{\langle |\nabla h|^2 \rangle} = \sqrt{\left\langle \left(\frac{\partial h}{\partial x}\right)^2 + \left(\frac{\partial h}{\partial y}\right)^2 \right\rangle} = \sqrt{2m_2} \tag{43}
$$

where the equality depends on the result that, for an isotropic surface, the orthogonal components $\frac{\partial h}{\partial x}$ and $\frac{\partial h}{\partial y}$ are uncorrelated.

The definition of RMS curvature $h''_{rms}$ is less common, but we shall follow PR [25] in defining

$$
\begin{aligned}
h''_{rms} &= \sqrt{\langle (\nabla^2 h)^2 \rangle} = \sqrt{\left\langle \left(\frac{\partial^2 h}{\partial x^2}\right)^2 + \left(\frac{\partial^2 h}{\partial y^2}\right)^2 + 2\left(\frac{\partial^2 h}{\partial x^2}\right)\left(\frac{\partial^2 h}{\partial y^2}\right) \right\rangle} \\
&= \sqrt{m_{40} + m_{04} + 2m_{22}} = \sqrt{8m_4/3}
\end{aligned}
\tag{44}
$$