On Pastewka & Robbins’ criterion for macroscopic adhesion of rough surfaces

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Abstract
Pastewka & Robbins (PNAS, 111(9), 3298-3303, 2014) recently have proposed a criterion to distinguish when two surfaces will stick together or not, and suggested it shows a large conflict with asperity theories. It is found that their criterion corresponds very closely to the Fuller and Tabor asperity model one when bandwidth $\alpha$ is small, but otherwise involves a rms amplitude of roughness reduced by a factor $\sqrt{\alpha}$. Therefore, it implies the stickiness of any rough surface is the same as that of the surface where practically all wavelength components of roughness are removed except the very fine ones, which is perhaps counterintuitive. The results are therefore very interesting, if confirmed. Possible sources of approximations are indicated, and a significant error is found in plotting the pull-off data which may improve the fit with Fuller and Tabor. However, still they show finite pull-off values in cases where both their own criterion and an asperity based one seem to suggest non stickiness, and the results are in these respects inconclusive.

Keywords: Adhesion, Greenwood-Williamson’s theory, rough surfaces

1 Introduction
Pastewka & Robbins (2014, PR in the following) recently suggested a criterion to distinguish when two surfaces will stick together (i.e. when the area-load curve bends into the tensile quadrant), which seems based only on fine scale quantities like rms slopes or curvatures, and argued that it conflicts with the classical criterion obtained by Fuller & Tabor (1975, FT in the following) using an asperity model, where instead emphasis is on rms amplitude, both for stickiness and for the value of pull-off. With beautiful atomistics simulations, PR introduce self-affine fractal roughness from a lower wavelength $\lambda_s$ of order
nanometers $\lambda_s/a_0 = 4, 8, 32, 64$, to an upper wavelength $\lambda_L$ in the micrometer
to millimeter range, e.g. $\lambda_L = 2048a_0$, where $a_0$ is atomic spacing.

Their main initial experiment is described as varying the level of adhesion
and adjusting the external load $N = N_{rep} - N_{att}$ as to keep constant the repulsive
contact area. They find that:

1) There is always a linear relation between the external load $N$ and the
area in intimate repulsive contact, $A_{rep}$. A result that was shown to be robust
in asperity models and was not questioned until people started to be interested
in very large bandwidths roughness. Defining Nayak bandwidth parameter
$\alpha = \frac{m_2}{m_0^2} \sim \frac{\zeta^{2H}}{\zeta}$, where $m_n$ are the moments of order $n$ in the random process,
$\zeta = \lambda_L/\lambda_s$ is magnification factor, and $H$ is Hurst exponent, PR systems have
for the low fractal dimensions ($H = 0.8$) a Nayak $\alpha$ of the order of 1600, which
is very large, and at these large bandwidths asperities coalesce and form bigger
objects which are difficult to be defined by random process theory (Greenwood,
2007). This leads in asperity models to an area-slope which is linear only asymptotically at large separations, and decreasing with $\alpha^{1/4}$ otherwise (Carbone &
Bottiglione, 2008); but let us not distract the reader with this point which,
in the asperity adhesive models, may tend to decrease stickiness, whereas we
shall see that PR criterion introduces a bandwidth dependence which strongly
increases stickiness, and with $\alpha^{1/2}$.

2) They find the attractive forces have little effect on the detailed morphology
of the repulsive contact area, suggesting the corresponding repulsive force
and mean pressure are also nearly unchanged. This suggests they are close to
the Derjaguin-Muller-Toporov (DMT) limit for which the repulsive pressure is
unaffected by adhesive forces and hence the deformation is principally due to
the repulsive forces, which in the DMT theory is given by Hertz theory.

3) They notice that in the ”attractive” regions, the pressure is simply the
theoretical strength of the material, $\sigma_{th} = w/\Delta r$, where $w$ is surface energy, and
$\Delta r$ is a range of attraction. This suggests a sort of Dugdale-Maugis model for
adhesion which requires only the knowledge of the size of the region of attractive
forces, $A_{att}$. $A_{att}$ is found to be a fixed proportion of the repulsive one $A_{rep}$,
by considering the first order expansion of the separation distance between two
contacting bodies under repulsive forces only, which scales as distance$^{3/2}$, and
equating the peak separation to the characteristic distance $\Delta r$. Notice in particular both $A_{att}$,$A_{rep}$ are written as a function of a perimeter $P$, respectively

$A_{rep} = Pd_{rep}/\pi$ and $A_{att} = Pd_{att}$, where $d_{rep}$ and $d_{att}$ are the characteristic
contact diameter and the additional size of attractive region, respectively, sug-
gest the contact area is a ”fractal”, which requires special attention. However,
at least in the limit of low bandwidths, the simpler model of circular contact areas of diameter $d_{rep}$, and circular annuli $d_{att}$ around the repulsive contact
areas, should be sufficient. An asperity model would also show this if it pre-

1With this definition of $d_{rep}$, which is the mean over contiguous segments in horizontal or
vertical slices through $A_{rep}$, for a set of $n$ circular objects, we get $A_{rep} = nd_{rep}^2$ instead of
$n\frac{\pi}{4}d_{rep}^2$, which means that the representative diameter is a little smaller than the real one,
$\bar{d}_{rep} = \frac{\sqrt{\pi}}{2}d$.  

2
dicts the repulsive and adhesive loads to be proportional each to the number of asperities in contact, \( n \). This will be shown to be indeed the case. In other words, the perimeter \( P \) can be given by \( n\pi d_{rep} \) and the entire set of results continues to hold for the asperity model too. For the circular area case, in particular, the PR calculation leads to a circular attractive annulus of size \( d_{att} = \frac{1}{d_{rep}} \left( \frac{2}{3} R \Delta r \right)^{2/3} \) and an attractive load per asperity

\[
N_{att,asp} = \pi d_{rep} d_{att} \frac{w}{\Delta r} = 3^{2/3} \pi w R \left( \frac{\delta}{\Delta r} \right)^{1/3}
\]

where \( \delta \) is the compression of the asperity, suggesting this model doesn’t lead exactly to the DMT model for a sphere (see Maugis, 2000) as usually it is reported that for DMT the adhesive load on the asperity is independent on its compression and is equal to \( N_{att,asp} = 2\pi Rw \), the pull off load. However, this point doesn’t change the main results of this discussion, and we shall take the PR model for the calculation of the asperity theory, rather than the original DMT.

4) A condition for stickiness is found in their eqt.10

\[
\frac{h'_{rms} \Delta r}{\kappa_{rep} l_a} \left[ \frac{h'_{rms} d_{rep}}{4\Delta r} \right]^{2/3} < 1
\]

In loose terms, PR criterion says nothing new: that for macroscopic bulk solids, adhesion at the macroscale is observed only in the case of very soft bodies of very smooth and clean surfaces, so that the length scale \( l_a = w/E^* \) is sufficiently large compared to \( a_0 \), where \( E^* \) is plane strain elastic modulus of the material pairs, and \( a_0 \) is atomic spacing. More precisely, there is a limit in vacuum for perfectly clear surfaces of crystalline solids, \( l_a/a_0 \approx 0.05 \) for a Lennard-Jones potential whose interaction distance \( \Delta r \approx a_0 \). However, it is the detail that matters. Using well established results \( \kappa_{rep} \approx 2, \Delta r \approx a_0 \) but grouping the variables using the Nayak bandwidth parameter, we can restate (2) as

\[
\frac{h'_{rms} a_0}{2l_a} \left[ \frac{h_{rms}}{a_0 \sqrt{\alpha}} \right]^{2/3} < 1
\]

and therefore really the condition is on rms amplitude also for PR. Despite this condition does not correspond immediately to the original FT parameter (which contains a radius of asperities), we shall find that a very close equation is obtained also with very simple asperity models, except that the \( \sqrt{\alpha} \) reduction of \( h_{rms} \) is not obtained, which means that asperity models predict a much stronger reduction of stickiness with roughness amplitude.

2 A simple asperity model

We can restate the basic results of the FT model in a simpler form if we consider some simplified assumptions, without changing the results qualitatively. We
consider therefore an exponential distribution $\phi = \frac{C}{s} \exp \left( -\frac{s}{\sigma_s} \right)$ ($s > 0$), and use the PR model for the behavior of each of the asperities [11], namely the adhesive load on each asperity of radius $R$ is dependent on compression $\delta$ with a power-law. Repeating the standard calculation of asperity models (see Johnson, 1985), and the contact area as being purely given by the compressive actions, the number of asperities (per unit area) in contact $n$, and the total area $A$ are unchanged with respect to the standard Hertzian case without adhesion,

$$ n = D_0 \exp \left( -\frac{d_0}{\sigma_s} \right) \tag{4} $$

$$ A_{rep}/A_0 = \pi R \sigma_s n \tag{5} $$

where $D_0$ is total number of asperities per unit area. The total load per unit area is instead changed as

$$ N/A_0 = n \left( E (\sigma_s^2 R)^{1/2} \sqrt{\pi} - 3^{2/3} \Gamma \left( \frac{4}{3} \right) \pi w R \sigma_s^{1/3} \Delta r_{1/3} \right) \tag{6} $$

PR suggest a critical importance of geometry of the contact not being "euclidean", but being fractal. They find the contact area as a intricate geometry having a characteristic size which they estimate from purely geometrical considerations

$$ d_{rep} = 4 h_{rms}'/h_{rms}'' \tag{7} $$

which has to be multiplied by a perimeter, where the dependence on the contact load enters. We try to reinterpret this result in the light of asperity model maintaining circular contact areas, and simply stating that the perimeter varies with number of asperities in contact, and is therefore a multiple of $d_{rep}$ itself. Dividing (5) by (4), we have an estimate of the mean diameter for the asperity model

$$ d_{rep,am} = 2 \sqrt{R \sigma_s} \tag{8} $$

and hence seems to be dependent on non-local quantities, in contrast with (7). However, using well known quantities in random process theories (see Carbone & Bottiglione, 2008) for the product $R \sigma_s D_0 = \frac{1}{48} \sqrt{\frac{3}{\pi}} (\alpha - 0.9)$ which was in early days considered to be constant, but which instead varies with bandwidth, and for $D_0 = \frac{1}{6 \pi \sqrt{\frac{m_2}{m_4}}}$, we get

$$ d_{rep,am} = 2 \left( \frac{6 \pi \sqrt{3}}{48} \sqrt{\frac{3}{\pi}} (\alpha - 0.9) \sqrt{\frac{m_2}{m_4}} \right) = (3.2 \div 10.3) h_{rms}'/h_{rms}'' \tag{9} $$

changing bandwidth in the range used by PR (16 to 1600), so this evaluation gives radius generally higher than PR finds. Exact coincidence occurs only for $\alpha \simeq 40$. This is still a correct order of magnitude result with respect to PR calculation, and indeed PR suggest that their factor 4 is an estimate "deviations
by up to a factor of 2 from this expression for $d_{rep}$ are responsible for the spread in the figure 3, but should we attribute the scatter to a bandwidth dependence as the asperity model predicts? It is extremely important as this assumption changes quite radically the result on the stickiness parameter. Indeed, they also suggest "For a given system, changes in $d_{rep}$ with $A_{rep}$ are less than 25% over 2–3 decades in $A_{rep}$." Since for a given system implies a given bandwidth, PR also find indirectly that the most part of the variation is due to bandwidth, and the factor 2 they find seems surprisingly in agreement with our estimate for (9) which is indeed a factor 2 larger for large bandwidths. If we were to modify their criterion (3) with this $\alpha^{1/4}$ increase of $d_{rep}$ with bandwidth in (9), we would already restrict their result as

$$\frac{h_{rms}a_0}{2l_a} \left[ \frac{h_{rms}}{a_0\alpha^{1/4}} \right]^{2/3} < 1$$

Returning to the load equation (6), it results from a difference, and hence it becomes zero when the contact becomes "sticky". To compare with more advanced random process theory based asperity models (see e.g. Carbone & Bottiglione, 2008), the term $\sqrt{\frac{R}{\sigma_s}}$ transforms into a slope parameter (we are confusing of course $\sigma_s$ to a rms amplitude), and therefore there is a sharp distinction between sticky and nonsticky behaviour when

$$\chi = 0.33 \frac{a_0}{l_a} h_{rms} \left( \frac{h_{rms}}{a_0} \right)^{2/3} < 1$$

which is remarkably close both qualitatively and quantitatively to PR parameter (3) at low bandwidths: exact coincidence would be obtained for a special bandwidth parameter, which in our crude estimate is of the order of $\alpha = 3.5$.

PR criterion also suggests that, if we consider a full self-affine spectrum of roughness, since the rms slopes and curvatures are defined only by the fine scale features, if these fine scales satisfy the criterion, it does not matter if we have this fine roughness structure as part of a much wider bandwidth of roughness, or in itself. In other words, if we start of with a fine roughness structure so that $\alpha_{fine} = 2$ (fig.1b) then we can enlarge the roughness without limit if $h_{rms, big} = \sqrt{\alpha_{big} h_{rms, fine}^2}$ as in Fig.1a.

Notice that in a sense, this "removal" of large scale roughness was done by FT in a much crude way in the sense that they had a macroscopic form, and microscopic roughness, although it is unclear how many scales of roughness they had in the microscopic scale. In their comparison with experiments, they used the reduction of pull-off with respect to the case of aligned asperities in a case like Fig.1b, for their spheres. When they did compute the adhesion parameter, they only considered rms amplitude of the fine scale roughness: however, they used this reduction factor to correct the pull-off value expected for the spheres.

\[2\] In the FT model, the transition is not so sharp, but at low enough, the pull-off is so small and the region of negative loads is obtained at so high separations, that we can consider the cases non-sticky.
which scales with their radius. Hence, it would seem that in the more complex problem with multiscale roughness, if PR criterion is correct, we expect that pull-off cannot be dependent only on this new adhesion parameter.

3 Pull-off

PR have also interesting data for pull-off in their "Supplementary Information", which they find in error with respect to the FT prediction by several orders of magnitude and also qualitatively not in good order. First, we should note an error in the scale of their Fig.S3. PR were aiming at using the scale used by FT, the ratio of pull-off load to sum of pull-off of aligned total number

Fig.1 An example of the "stickiness" equivalence in PR criterion (a) a local fine scale roughness, on a larger wavelength structure of which we show only some parts, (b) the same roughness but now in itself.
of asperities. They assumed $R \sigma_s D_0 = 0.05$ which was correct in the old days for low bandwidths (it is $\frac{2}{38} \sqrt{\frac{2}{\pi}} (\alpha - 0.9)$, which is 0.05 only for $\alpha = 7$) whereas they bandwidth spans the range $\alpha = 16 - 1600$. So, if we keep their points as they are, we should have many curves for FT, spanning a band. For the largest bandwidths, the FT curves would be almost 2 orders of magnitude higher. Some points may still too "sticky" than what FT predicts, and especially "stickiness" results for a much wider range than the original FT adhesion parameter, in agreement with the main difference we found in the stickiness parameters. However, the reduction on rms amplitude needed to collapse the data in the x-axis is at most a factor 2, whereas the new PR criterion suggests a much higher reduction, scaling with $\sqrt{\alpha}$. PR suggest that their data are the "lower bound" of pull-off forces they can find, since these are load-dependent. Would other pull-off forces be closer to a FT theory "corrected" with the new adhesion parameter? It is impossible without estimating all individual bandwidths in the data.

However, yet another contradiction appears from the data: the caption says $h_{\text{rms}}' = 0.1$ or 0.3 (closed and open symbols), $l_a/a_0 = 0.005$ (blue) or $l_a/a_0 = 0.05$ (red). Hence, their own criterion now reads with $h_{\text{rms}}' = 0.1$ and the case with low adhesion $l_a/a_0 = 0.005$, suppose with $\lambda_S = 4a_0$, and $\alpha = 1600$

$$\frac{h_{\text{rms}}}{a_0} < \left( \frac{1}{10} \right)^{3/2} \sqrt{1600} = 1.26!$$

and by no means their surfaces are so small in rms amplitude to be of atomic size. Indeed, the rms amplitude they have can be estimated in this case to be $h_{\text{rms}} = h_{\text{rms}}' \lambda_s \lambda_L/\lambda_s$ $H = 0.1 \times 4a_0 (1000)^{0.8} = 100a_0$. Even more absurd with $\lambda_S = 64a_0$, bandwidth 74, still with same parameters, PR criterion reads $\frac{h_{\text{rms}}}{a_0} < \left( \frac{1}{10} \right)^{3/2} \sqrt{74} = 0.27$. If we take now $l_a/a_0 = 0.05$, these numbers will be multiplied by 10, which doesn’t solve the problem: most point in the plot should be non-sticky both for their criterion and an asperity based one. These pull-off values correspond, in the correct scale, to the pull-off of a relevant number of asperities out of the total number, and do not seem to be plausible with the parameters of roughness they have.

### 4 Discussion

We have pointed out that the "parameter-free" theory of PR may contain several important approximations which affect the stickiness criterion. In particular, their assumption of a constant factor 4 in $d_{crp}$ seems problematic even in fig.3 PR show, and conflicts by the same factor as we have estimated in an asperity model. The fact that asperity models at low bandwidths correctly describe the geometry of the problem is well accepted today (Greenwood, 2007), so the source of conflicts seems to be the dependence on $\alpha$ for large bandwidth.

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3PR use $\frac{3}{2} \pi R \sigma$ for a single asperity as in JKR theory, instead of DMT value which may be more appropriate, but this is irrelevant.
Another difference with the asperity model is hidden in assuming mean values for both diameter of repulsive contact area and size of annulus of attraction. In PR parameter-free theory, $d_{rep}$ is the mean diameter and does seem to take into account of the distribution of contact spot sizes, and so does $d_{att}$. In fact, the asperity model does not need to make this approximation. If I estimate the mean size of the annulus of attraction directly from the $d_{rep,am}$ in (9), as $d_{att} = \frac{1}{d_{rep}} \left( \frac{4}{3} R \Delta r \right)^{2/3}$, I get

$$
\frac{A_{rep}}{A_{att}} = \frac{n\pi d_{rep,am}^2 / 4}{n\pi d_{rep,am} d_{att}} = \frac{(\sigma_s / a_0)^{2/3}}{4 \left(\frac{3}{2}\right)^{2/3}} \approx \frac{(\sigma_s / a_0)^{2/3}}{5.24}
$$

(13)

whereas if I estimate $\frac{A_{rep}}{A_{att}}$ from the full integration process which takes into account of the distribution of contact spots sizes (6) as

$$
\frac{A_{rep}}{A_{att}} = \frac{N_{att}/A_{rep}}{w/a_0} = \frac{(\sigma_s / a_0)^{2/3}}{3^{2/3} \Gamma \left( \frac{4}{3} \right)} \frac{1}{1.85}
$$

(14)

which suggests a 3 times less area of attraction. As $\sigma^{1/4}$ varies from $16^{1/4} = 2.0$ to $1600^{1/4} = 6.3$, this factor 3 is not irrelevant. It may well be that these subtle differences in the factor are better captured by the PR model instead of the asperity model, but the result seems quite counterintuitive.

5 Conclusion

We have reexamined the results of PR recent "parameter-free" theory. The parameter-free theory in fact does contain some parameters, and in particular, the estimate of the diameter of the repulsive contact areas, which deserves further attention. Despite asperity theories are known to be possibly in error at large bandwidth parameters, many results PR find numerically do not seem in conflict, except of course the criterion for stickiness, which corresponds only in the limit of low bandwidths. The new criterion contains a curious implication, that one can take some fine scale roughness, and build on it increasingly larger wavelengths of roughness without affecting the stickiness. Since a finite stickiness implies also a finite pull-off, this seems to be an interesting result, which requires further proof. Unfortunately, the data they present for pull-off do not seem consistent, and do not permit conclusive discussion.

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