

CONTACT & ADHESION AT ATOMIC SCALES¹

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In two interesting papers, Luan & Robbins have studied the effect on molecular roughness on the contact mechanics at atomic scale, typical of SPM tips. They observe large differences in the numerical results of MD simulations with respect to the theory corresponding to the smooth single asperity spherical tip, i.e. neglecting roughness, namely Hertz or JKR (or Maugis-Dugdale). We suggest Luan & Robbins' results can be understood using the asperity model of Greenwood & Tripp instead of Hertz (for the case with no adhesion), and the asperity model of Fuller & Tabor, instead of JKR or Maugis-Dugdale (for the adhesive case). The latter comparison is only very qualitative, since the equivalent of Greenwood and Tripp for the adhesive case has not so far been developed (unfortunately). Further remarks are devoted to stiffness, and the analogy to thermal-electrical conductance. Finally, it is remarked that most theories predict expected mean values of the results as a function of roughness, and no theory presently is able predict the scatter in results due to roughness, and that also would be most useful.

1. Introduction

Luan & Robbins recently published two very interesting papers, the first on Nature (2005), and the second on PRE (2006) (we shall refer in the following as L&R05, and L&R06, respectively). They remarked that traditional calculations using continuum contact mechanics can be applied at ever smaller lengths (although they are known to break down at atomic dimensions), to fulfil the interest in nano-devices, or studying the molecular origins of friction and adhesion. However, atomic-scale surface roughness is always produced in practise (except for a bent perfect crystal) and leads to deviations from continuum theory which for contact areas and stresses may be a factor of two, and for friction and lateral contact stiffness may even be an order of magnitude. We shall show however that simple calculations using continuum models for roughness also justify such deviations. Naturally, the original motivation of the L&R papers was to assess the validity of continuum mechanics in interpreting results of experiments using scanning probe microscopes (SPM) whose roughness is not well known² and hence the use of asperity theories including roughness is not a viable alternative, and the L&R message remains that difficulties or errors may result from too simple analysis of the experimental data not considering molecular roughness.

2. Contact mechanics (no adhesion).

For the surfaces shown in Fig.1 of L&R05, the results in Figs.2,3 & S2 are much as we would expect from continuum contact mechanics. The displacement and contact area follow Hertz well and the stepped surface has a pressure distribution which could be predicted from the superposition of flat punches. The pressure distributions for the amorphous surface looks much like the computer calculations of normal scale elastic rough surfaces. The Greenwood & Tripp (1967) analysis of rough spherical surfaces, summarised in Ch.13 of KL Johnson's book (1985), shows that the effect of roughness on Hertz stress is principally governed by the ratio: $\alpha = rms / \delta$, where rms is the rms roughness and δ is the elastic compression at the load in question. From Fig.1b we estimate $rms \cong \sigma / 3$, where σ is the effective molecular radius. This gives $\alpha = \sigma / 3\delta$. At the two loads illustrated in Fig.3 $N/IRE^* = 0.0019$ & 0.0077 , and $\alpha \cong 0.2$ & 0.6 . These values are not inconsistent with Figs.13.12 & 13 in KL Johnson's book (1985).

¹ We acknowledge useful discussions with Ken Johnson (Cambridge) on this matter.

² Typically, SPM tips measure total forces and displacements with tips of radius $R < 10-100$ nm, and Luan and Robbins focus in particular on a radius typical of SPM tips, $R = 100\sigma$ i.e. 30 nm, where σ is the atomic spacing.

3. Adhesion.

This is perhaps a little harder problem for using rough models, because to date there is no model equivalent to Greenwood & Tripp (1967) including adhesion. Fuller and Tabor's theory (1975) on adhesion of a rough surface use the asperities model of Greenwood and Williamson (1966), with the JKR (Johnson, Kendall and Roberts, 1971) theory for adhesion of each independent asperity, to develop a theory *but for a nominally flat surface* having a Gaussian distribution of asperities. They introduce a “rough adhesion” parameter ϕ , which is the ratio of the

rms roughness to the elastic compression at pull off, $\phi = rms/\delta_c$, where³ $\delta_c = \frac{3}{4} \left(\frac{\pi^2 \cdot w^2 r}{E^*} \right)^{1/3}$, where

w is work of adhesion per unit area (commonly denoted in terms of the adhesion surface energies $\Delta\gamma = \gamma_1 + \gamma_2 - \gamma_{12}$), and E^* is the combined elastic modulus. We shall refer to “ r ” as the radius of the asperity and use the capital letter “ R ” to later identifying the macroscopic radius of the sphere.

F&T obtain that the reduction of the pull-off force with respect to the force needed to peel all asperities nominally in contact is a function of this parameter only. For example, for $\phi=0.75$ there is a reduction of about 50%, whereas a value of 3 corresponds to almost complete loss of adhesion. We write ϕ here in the reciprocal form

$$1/\phi = 4/(3 \pi^{2/3}) r^{1/3} w^{2/3} / (E^{*2/3} rms) \quad (1)$$

We introduce now the parameter introduced by Tabor (1977) to show the two theories for adhesion of a single asperity spherical contact, the JKR and the DMT (Derjaguin, Muller, Toporov, 1975) applied to opposite extremes of the range (low and high, respectively)

$$\mu_R = R^{1/3} w^{2/3} / (E^{*2/3} z_0) \quad (2)$$

where we have used the notation μ_R to remind that this is relative to R , the reduced radius of the macroscopic spheres (if there are two spheres in contact), and z_0 is the equilibrium separation (for example in the Lennard-Jones potential). In most materials, both z_0 and the ratio w/E^* are of order 1 nm. Hence, the JKR limit is only reached by increasing R to macroscopic dimensions of micrometers or larger. Even in the simulations of Luan and Robbins μ lies between 0.1 to 0.75 i.e. between JKR and DMT limits (more precisely, the transition could also involve the theoretical strength, see Gao and Yao, 2005). For this reason, they compare their results to the Maugis-Dudgale theory (Maugis, 1992). Now, for exactly the same reasons, since r the radius of the “asperity” is very small, $1/\phi$ which resembles the “local” Tabor parameter for the asperity, is certainly very small, i.e. ϕ is quite large. In other words, from the one hand, we cannot use JKR for the local asperity, on the other hand, we expect the effect of local “asperities” to be possible very large, in “destroying” adhesion.

More precisely,

$$\phi = 1.608 (R/r)^{1/3} (rms/z_0) (1/\mu_R) \quad (3)$$

As before, the ratio rms/z_0 is hard to quantify precisely, but from Fig.1b of in L&R05 we estimate $rms \cong \sigma/3$, and from L&R06 we know σ/z_0 to be 2, so that it is clear for the amorphous surface of

³ This can also be written as $\delta_c = \left(\frac{P_c^2}{3K^2 r} \right)^{1/3}$ where $K = \frac{4}{3} \frac{E}{1-\nu^2}$, and P_c is the pull-off force for the JKR model

$$P_c = \frac{3}{2} \pi w \cdot r.$$

Fig.1b that they are of the same order, so we will take 2/3. The ratio (R / r) is even more difficult to quantify (and clearly it would not be the same for the stepped surface, for which it is not even clearly defined), but as a cockshy take r to be 3σ (also because an asperity of less than 2 atoms is difficult to imagine!), and considering R/ σ is said to be 100, and gives

$$\phi = 1.608 \cdot 3.2 \cdot 0.66 / \mu_R = 3.58 / \mu_R \quad (4)$$

For an AFM tip, values of μ have been found in the range 0.2 - 0.8 (indeed L&R give 0.1 to 0.75), suggesting ϕ in the very high range 18 - 5 respectively. Referring to the Fuller & Tabor plot, this corresponds to *complete loss of adhesion*! Even in the JKR regime where $\mu > 6$, say, $\phi < 0.5$, for which the effect on adhesion is still not negligible. In effect, for adhesion, L&R06 report that results for bent tips could be fit with M-D theory, but the values for stepped and amorphous tips lie well outside M-D theory, showing factor of 2 deviations for stepped tips, and in general change by factors of 2 to 4. This are entirely compatible to a F&T interpretation.

One problem is that, when we apply F&T theory, we are comparing the effect of adhesion with respect to the pull-off force of the total nominal number of asperities in contact, which is not easy to quantify with respect to the pull-off force of the macroscopic tip. For that, a model combining F&T with Greenwood and Tripp would be needed. Such a model, having adhesive asperities on top of a sphere would clearly show, in analogy to Greenwood and Tripp, that the effect of roughness on pull-off force is principally governed by the ratio:

$$\phi = rms / \Delta_c, \quad (5)$$

where now Δ_c is the elastic compression at the pull off load *for the macroscopic sphere*. The F&T parameter $\phi = rms/\delta_c$, viceversa, correctly uses only geometrical features of the asperities, since their model contains only a distribution of asperities and no macroscopic scale of the contact. Obviously, $\phi = rms/\delta_c$ and $\phi = rms / \Delta_c$, mean that since $\delta_c \ll \Delta_c$, and precisely $\delta_c/\Delta_c = (r/R)^{1/3}$ then $\phi \gg \phi$, and therefore it is possible that, although F&T suggests a big effect on the pull-off force, a complete theory would suggest a lot less sensible effects. Naturally, the use of F&T or even this mode statistical advanced theory for this kind of condition where only a few asperities are in contact is very unlikely to be quantitative, and one wonders if a simple strength controlled limit is reached for many cases.

In spite of the above calculations it must be bourn in mind that AFM tips have often been observed to show strong adhesion and approximately JKR behaviour. This is at least partly paradoxical and perhaps needs further investigation. Partly, this could be an effect of increased adhesion in asperities with respect to the JKR model, as shown by the extension of Fuller and Tabor to deal with the Maugis-Dugdale model (Morrow et al. 2003), or to the DMT model (Maugis, 1996), but more likely the effect of full contact is not modelled by asperity models, and truly multiscale models (at least, two-scales as Greenwood and Tripp), not using JKR as the local asperity constitutive equation, would be interesting.

4. Stiffness.

That stiffness exacerbates the effects of roughness was well know, since it is a derivative of the load-displacement relation, and clear the presence of an asperity completely changes the curve. Incidentally, an important connections between elastic stiffness and thermal and electrical conductance is given the analogy between conductance and incremental stiffness of a contact (Barber, 2003) --- indeed the electrical conductance C at any load is proportional to the incremental stiffness in the elastic contact problem, the derivative of the load P with respect to the separation d

$$C = -\frac{2}{\rho E} \frac{\partial P}{\partial d} \quad (6)$$

where E is combined elastic modulus and ρ is the combined resistivity of the materials. In the case of a nominally flat surface, it is easy to show that the stiffness for the Greenwood-Williamson model is simply linear with total load, and inversely proportional to the “amplitude” of the parameter σ_s for the simplified case of exponential distribution of asperities roughness,

$$S_{GW \exp} = \frac{P}{\sigma_s} \quad (6)$$

showing that the only asperity feature which seems to matter for the electrical conductance is a parameter related to amplitude of the heights (and not the slopes and curvatures). However this result, based on a smooth continuous distribution of identical asperities, hides the true response in numerical experiments. For example, in Ciavarella et al (2006a), various Weierstrass –Mandelbrot fractal surfaces are generated, and the stiffness (or conductance) –load relationship is vice versa only approximately given by the original GW theoretical model. Significant deviations from linearity and significant scatter seem to be found, particularly at low fractal dimensions and large separations. Effect of interaction is seen strongly at low separations, where scatter is minimal. This is better seen in (Ciavarella et al 2007) with a new formula to consider the mean interaction effect in the Greenwood and Williamson model, which is equivalent to an increase of the effective separation of the mean planes by a quantity proportional to the nominal pressure, resulting in a reduction of the “real” area of contact and of total load for a given separation. A parallel is drawn between this correction and the “clustering” terms of resistance in the Holm-Greenwood formulae for a cluster of circular spots. In the presence of a macroscopic shape, of course there is a further effect which is of the “series” stiffness of the macroscopic shape itself.

Turning back to (Barber, 2003), bounds are derived based on the idea that bounded amplitudes of roughness cannot “displace” the load-displacement curve by more than the size of the roughness itself. Qualitatively, all these effects are entirely compatible to the Luan and Robbins results.

5. Conclusions

Theories on asperity contacts, and in general rough contact, help understanding the apparently surprising results of Luan and Robbins on molecular roughness of SFM tips. In fact, at loading and geometrical conditions such that the roughness is comparable to the elastic compression (the parameter in Greenwood and Tripp $\alpha \cong 0.2$ & 0.6) the contact is of course bound to deviate from that of a smooth sphere. Similarly, for the adhesive case, and particularly in terms of stiffness. When dealing with macroscopic bodies, we were able to judge the effect of roughness more simply, since we knew that the effect of microns of roughness could be only sensible at microns of induced elastic compression. It is however also true that the effect of roughness in macroscopic bodies is more subtle than what we think at first sight. Indeed, if it wasn't for roughness, the majority of the laws governing tribology and its empirical laws would be completely different. We would probably have a lot more adhesion around, the friction would not be to the first order simply proportional to normal load (via a “friction coefficient”), and so on. Hence, it is interesting that we now show surprise if roughness has a role even into the nano “scale” of tribology. Of course, here even nanoscopic amounts of roughness (indeed molecular roughness due to the exact arrangement of atoms) plays a role. The theories of elastic contact with roughness, perhaps in the simplest form using asperities, are very useful, and missing ingredients we identify here are (i) the Greenwood and Tripp equivalent or a full multiscale version of asperity model with adhesion, not limited to the JKR regime of course, (ii) a model of scatter, i.e. capable of predicting not only the mean values, as all theories today, but also the expected amount of scatter in the results. We have deliberately not discussed the case of friction, since it would lead us too far for this small note.

6. References

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