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Discussion of "Measuring and Understanding Contact Area at the Nanoscale: A Review" by Tevis D. B. Jacobs and Ashlie Martini

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Jacobs and Martini (JM in the following) give a nice review of direct measurement methods (in situ electron microscopy), as well as indirect methods (which are based on contact resistance, contact stiffness, lateral forces, and topography) for measurement of the contact area, mostly at nanoscale. They also discuss simulation techniques and theories from single-contact continuum mechanics, to multi-contact continuum mechanics and atomistic accounting. As they recognize, even at very small scales, "multiple-contacts" case occurs, and a returning problem is that the "real contact area" is often an ill-defined, "magnification" dependent quantity. The problem remains to introduce a truncation to the fractal roughness process, what was called in the 1970's "functional filtering". The truncation can be "atomic roughness", or can be due to adhesion, or could be the resolution of the measuring instrument. Obviously, this also means that the strength (hardness) at the nanoscale is ill-defined. Of course, it is perfectly reasonable to fix the "magnification" and observe the dependence of contact area, and strength, on any other variable (speed, temperature, time, etc).

1. Introduction

One of the basic laws of friction dictate *"force of friction is independent of the apparent area of contact"* and was already noticed by Leonardo da Vinci (Pitenis et al 2014). The geometry of Leonardo's experiments is still of interest after 500 years, since we are still trying to understand what happens to the "contact area" by measuring it with a different resolutions. JM's interest is to look at the possibility of quantitatively measuring the real contact area via indirect measurement of other quantities, like friction force, or thermal or electrical conductance. This probably originates from Bowden and Tabor (1939) (BT in the following) who first stated, "*some knowledge of the real area of contact is essential for any complete understanding of the mechanism of friction*". This statement seems to have inspired many researchers but perhaps BT were naïve or could not imagine how complex this "knowledge" of the contact area could be. BT also remarked that "*optical methods cannot reveal irregularities much smaller than half a wave-length of light*", and therefore already suggested that some measurements are really truncated at "mesoscale". BT model was mainly plastic, and their electrical conductance and visual measurements led them to suggest the real contact area to be *"less than one ten-thousandth of the apparent area"*. After many years, this conclusion appears vaguely correct, but probably it would be hard to have even today a precise

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number for "real contact area". JM review is after all still about this problem. Now, in many cases the "real contact area" is a well defined quantity, like when we can clearly identify a single contact. This occurs generally for soft and perhaps sticky solids, or at extremely small scales. But even at nanoscales, as JM review, roughness cannot be neglected. We take therefore the liberty to make some remarks to this problem, complementary to the review of JM, with the aim to stimulate even more debate: is there in fact a risk that, after very large effort, the real contact area is a quantity ill-defined, and therefore of limited interest?

2. Archard model for rough contact

Archard (1957) introduced the concept that, due to wear, plastic deformation on asperities cannot be going on forever. Archard made a step backwards with respect to the BT model, and attempted to justify Amonton's law (proportionality between normal and tangential load), via another mechanism alternative to plasticity. He then invented a "rudimental" fractal model of roughness, with his spheres on spheres on spheres on…, showing the real contact area tended to be linear with total load. But Archard did not observe that the coefficient of proportionality, in the limit, tends to zero, i.e. the contact area is a fractal itself! Amonton's law holds because the tangential force is proportional to the real contact area, which is zero times the normal load: shouldn't this lead to zero friction? It was only much later that Ciavarella et al (2000) remarked this, although with the not very popular choice of a Weierstrass series as a fractal, and Ciavarella and Demelio (2001), with the original Archard model. It did serve the purpose to show that "*no applied mean pressure is sufficiently large to ensure full contact and indeed there are not even any contact areas of finite dimension — the contact area consists of a set of fractal character for all values of the geometric and loading parameters*". The contact area was found to have a limiting fractal dimension of (2−D), where D is the fractal dimension of the surface profile. In other words, the higher the fractal dimension of the profile, the lower the fractal dimension of the contact area. Similar conclusion was obtained by Persson (2001), who introduced the idea of "magnification"-dependent solution, which is the another way to look at the "ill-defined" nature of the contact area.

3. Functional filtering

The problem of the "resolution-dependence" of the contact area had emerged even earlier. Greenwood-Williamson (1966) introduced the well known model of asperities (GW model), which simply assumed that radius of asperity would be "measured" by some instrument as some constant value. Later, Whitehouse & Archard (1970) (WA in the following) introduced the autocorrelation function (ACF), and discovered that their ground surface had an exponential ACF. They analyzed the implications of this: that whatever sampling interval they chose, between one-third and one-quarter of all their sample points would be a peak and that the mean peak curvature depended strongly on the sampling interval. Their measurements beautifully confirmed their predictions. WA noted that the Fourier transform of an exponential ACF was a power spectrum not too far from the accepted ones today (tending to power law at large wavevectors), but made nothing of this. In order to respond to this difficulty put forward by WA, in the 1970's an approach called "functional filtering" was advanced (Thomas and Sayles 1973, 1978) (TS in the following). It never became very popular, at least under this name, perhaps because each problem requires its specific recipe.

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Indeed, both long and short wavelength cutoff could be filtered, depending on "*the practical problem in hand*". It is remarkable to notice that TS referred to "non-stationarity of real surfaces", leading to fundamental parameters such as roughness and correlation length being also non-intrinsic --- this is still valid today, but it is generally forgotten.

All models predict the linearity of real contact area with load, starting from the plastic models of Bowden and Tabor, to the elastic Archard or Ciavarella et al.(2000)., or GW or Persson's theory. However, while we have a qualitative explanation of Amonton's law for friction, the coefficient of proportionality between area and load remains "resolution-dependent".

4. Can "adhesion" make the contact area well defined?

Another possible solution to the "contact area" ill-posed nature may come from adhesion. In their review, JM discuss the DMT (Derjaguin et al., 1975) and JKR solution (Johnson et al., 1971) for Hertzian contact which inevitably raise the question whether tensile stress has to be considered within the real contact area or not. "Contact" in the absence of adhesion (as JM remark) is defined by actually "hard-wall" contact and zero gap between bodies (by mathematicians, the "Signorini condition"). However, this is a simplification of a surface-force law that, in the absence of third bodies, should be based on the well-established Lennard-Jones "6-12" potential, which acts between pairs of molecules. This full LJ surface law leads however to a very complicated problem requiring a numerical solution. The earlier solution to this was the JKR model (Johnson et al., 1971) (originally developed for the Hertzian geometry) which turns out to be accurate for soft bodies and sufficiently large scale. It is not clear under which conditions this applies to rough bodies.

The discussion involving roughness is more recent. Asperity theories (Fuller and Tabor, 1975) seemed to have obtained the main result that a small rms amplitude of roughness destroys adhesion, also in comparison with experiments. However, the problem was not verified against the "fractal issue". Since there is also a dependence on radius of asperities, asperity models do not give a final "converged solution" (of the contact area) for a fractal. Pastewka-Robbins (2014) (PR in the following) have formulated a criterion for "stickiness" in the "DMT regime" by numerical observation of when the slope of the (repulsive) area-load becomes vertical which seems, surprisingly, independent on rms amplitude. This appears in contrast with asperity theories, and perhaps even with common experience. Pastewka-Robbins conclusions may be limited to the range of parameters they considered (see Ciavarella, 2016, 2017a, 2017b, Ciavarella and Papangelo, 2017): as rms slopes and curvatures do not converge for a true fractal, PR seem to suggest that everything is played at the upper truncation wavevector, i.e. at the atomic scale: is this realistic? For pull-off, which is a well defined macroscopic quantity, probably much better defined than the slope of the (repulsive) area-load, even PR observe a dependence on rms amplitudes, and their results are not qualitatively in contrast to asperity models. We are not sure if other qualitative conclusions of asperity models, like for example the sensitivity to the tails of the distribution (see Ciavarella et al., 2017) will be confirmed by more advanced simulations, considering that adhesion may be a lot more complex than what asperity models suggest. Notice that, in the limit of a rigid adhesion problem (Ciavarella & Afferrante 2016) an exclusive dependence on rms amplitude occurs, and it is therefore hard to think of the PR limit from this perspective.

Joe et al. (2017) generalize Persson's 2001 approach to Lennard-Jones adhesion, and suggest that the adhesive contact problem converges to a limit result when the spectrum is increased in fine details --- although this seems limited to the range when roughness increment have become of atomic scale. This is the first important indication of a real "regularization" of the contact problem, where slopes and curvature no longer matter and a proper solution for a fractal (without need to truncate the spectrum) can be defined.

In general, there is still no clear understanding of the possible regimes of adhesion of rough surface. The JKR limit seems also not clear: near full contact (Ciavarella, 2015, Ciavarella, M., Xu, Y., & Jackson, R. L. (2018)), it leads to a non-converging, paradoxical solution, actually sensitive to the 6^{th} order moment of the PSD. Clearly, the JKR assumption is in contrast with the limit at the smallest scales, and the adhesion problem shows itself much harder and richer than the adhesionless one.

5. Atomic roughness

If we have given too much emphasis to the ideal case of nominally flat surfaces, it is partly not our fault: as JM remark, the literature is much richer on this idealized case, and has neglected the probably much more important case of a tip with added roughness.

One of the few exceptions is Luan and Robbins (2005) (LR in following), who remarked that the atomic-scale surface roughness, always produced by discrete atoms in otherwise cylindrical surfaces, leads to dramatic deviations from continuum theory: *"Contact areas and stresses may be changed by a factor of two, whereas friction and lateral contact stiffness change by an order of magnitude. These variations are likely to affect continuum predictions for many macroscopic rough surfaces"*. For the surfaces shown in LR's Fig.1, the results in LR's 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 Ken Johnson book (Johnson, 1985) shows that the effect of roughness on Hertz stress is principally governed by the ratio: $\alpha_{GT} = h_{rms} /\delta$, where h_{rms} is the rms roughness and δ is the elastic compression at the load in question. From LR's Fig.1b we estimate h_{rms} / $\cong\sigma /3$, where σ is the effective molecular radius. This gives $\alpha_{GT} =\sigma /3\delta$. At the two loads illustrated in LR's Fig.3 we estimate $\alpha_{GT} \cong 0.2$ & 0.6. These values are not inconsistent with Figs,13.12 & 13 in Johnson book.

However, even Robbins himself in later studies (e.g. PR) *uses ideal roughness defined by self-affine processes on a rigid surface*, and not a multiscale roughness, which ends at real atomistic roughness like Luan and Robbins strongly seems to recommend. A matter of pure convenience to simplify the study?

6. Better posed and defined quantities

We don't always need to worry about the high-frequency cutoff, as some macroscopic quantities, stiffness, electrical and thermal conductance, for example, are well known to depend only on more macroscopic surface parameters, and principally on rms amplitude of roughness already since the

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times of asperity models. Not surprisingly, these quantities are often used to infer the contact area, as JM review.

In these respects, an important theorem was proved by Barber (Barber, 2003), showing that fine scale roughness cannot change the load-separation curve more than a certain amount. Hence, this remains true also for the stiffness or thermal and electrical conductance, which are connected by an analogy of the halfspace problems (under elastic assumption). Barber (2003) gives in other words also “bounds” to the stiffness, since the derivative of load-displacement curve also cannot change too much when adding roughness. But the bounds can be very “loose” if roughness is large.

In any case, the functional dependence of stiffness or electrical/thermal contact conductance is truly different from that of the contact area. The latter is a “resolution-dependent” quantity, and would not converge if we were to measure it with ever increasing resolution (or that it can only be defined when the resolution of the measuring instrument is defined). On the other hand, the stiffness is a true macroscopic quantity. In a model, it requires some degree of refinement, but it would converge to some well defined limit when we include more and more fine scales. How do we connect the two quantities, given this fundamental difference?

7. Fineberg’s expriments

We return to the “Leonardo” experiments as some spectacular recent measurement by the group of Jay Fineberg is perhaps appropriate in this context (Rubinstein et al., 2004, Ben-David and Fineberg 2011). Laser techniques with fast video recording permit today to measure the “net contact area”. This is obviously a “mesoscopic” contact area. Fineberg’s measurements have the somewhat specular goal to the papers JM mention: they measure the contact area to infer some information about the dependence of other quantities. In particular, a decrease of the contact area is qualitatively used to signal the occurrence of sliding. The basic Leonardo experiment appears richer and richer, as we enlarge our measurements capabilities, and our theoretical capabilities. This is a perfectly fine experiment, which has no particular need to define the “magnification” as there is no intention to “measure” quantitatively the contact area. Fineberg’s experiments show that detachment fronts transverse the interface at shear loads well below the full sliding critical value, as the detachment fronts change the real contact area at the interface by at least 20% (Rubinstein et al., 2004, Ben-David and Fineberg, 2011). Similarly, one can observe at this “resolution”, what happens to the contact area (or to the strength) as other quantities are varied, like temperature or speed, etc.

Conclusions

JM have made a good review of “nanoscale” contact, measurement, theory and simulation of contact area, and we have provided perhaps some additional comments. It is clear that a general “theory” for a real contact, be it nanoscale or macroscale, does not exist. Deviations from the classical standard Hertz theory or from rough contact theory can occur for many reasons, from type of shape, form and roughness, to friction to non-elastic behavior, to third bodies, to adhesion. Each of these reasons can involve very important complications and effects. The simulation of nanocontact itself is proving to be an area of some complexity, and simulation with molecular dynamics method are still far from being possible at macroscopic scales at realistic timescales. We have remarked that, already from the Archard model it can be inferred that the contact area, under elastic conditions, is in reality a fractal and hence is very “resolution-dependent”. This went

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perhaps unnoticed by Archard but was clearly remarked by Ciavarella et al.(2000). Despite improvements in accuracy over the classical GW theory, Persson's theory has not changed this fact. Quantities that depend on contact area are ill-defined. Load-separation curves and hence stiffness and contact resistance (thanks to Barber's analogy) are less ill-defined, and converge to some well defined value when more and more details of roughness are added: how do we compare with the "resolution-dependent" contact area? Atomic roughness is very rarely described accurately in models. Even multiscale models that are sensitive to truncation prefer to use a self-affine description truncated at some arbitrary point. Some inspiring recent results suggest that adhesion may be a "regularizing" ingredient to add. We perhaps conclude that to accurately and universally define the "true contact area" is a false goal: in most cases, we do not really need to know it. Perhaps in some cases, the measurement of the contact area at some "mesoscopic" scale is very useful in qualitative manner, as in the Fineberg "Leonardo" experiments.

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