

Chapter 2

Contact, Interactions, and Dynamics

E. Barthel

Abstract In this short introduction to tip–surface interaction, we focus on the impact of adhesion on the elastic contact of small spherical bodies. Standard notions are first reviewed but more complex contact conditions involving coatings or roughness are also considered. Special attention is devoted to dynamic response and ensuing dissipation.

2.1 Introduction: Contact and Adhesion

As the denomination suggests, in *force* microscopies, such as atomic force microscopy (AFM), ultrasonic force microscopy (UFM), etc. the interaction between the tip and the substrate lies at the core of the technique. Despite the A(tomic) in AFM, several atoms usually participate in the interaction, so that continuum scale approaches are relevant. The aim of this chapter is to explain some of the basic ideas underlying the adhesive contact of small objects like tips.

A distinctive feature here is the presence of curvature: one of the surfaces, the tip, is axisymmetric and curved, with radius of curvature R , so that the tip shape $f(r)$ is approximately

$$f(r) = \frac{r^2}{2R} \quad (2.1)$$

where r is the radial coordinate. Within the limitations of the following developments, this shape is also a good approximation to the local shape of a sphere, and for historical reasons we will often refer to the tip as the *sphere*. The other surface, the substrate, is flat.

E. Barthel (✉)
Surface du Verre et Interfaces, CNRS/Saint-Gobain,
UMR 125, 93330 Aubervilliers Cedex, France
e-mail: etienne.barthel@saint-gobain.com

In a basic view of the tip coming to the surface, the two surfaces initially sit at a separation distance d (Fig. 2.1a) along the symmetry axis and the gap between the surfaces is $d + f(r)$. When the surfaces are brought together, they come into contact as *point contact* and subsequently develop a *contact area* with finite size as the load increases (Fig. 2.1b). In terms of interactions, more or less long-range attractive interactions result in adhesion while short-range Born repulsion will provide for the contact side of the problem. It is the coupling of these interactions with continuum scale mechanical response which we consider here.

Sections 2.2, 2.3, 2.4 and 2.5 consist in an exposition of standard results for sphere contact mechanics while Sects. 2.6, 2.7 and 2.8 contain developments on the impact of more advanced features: coatings, roughness, and dissipation, keeping in mind some dynamic issues relevant for AFM.

The chapter has been designed for a reasonably straightforward reading. Beyond a mere enumeration of results, we also want to provide some hints as to the physical origin of the results. These details, and also more advanced ideas, which we believe would obstruct linear reading, appear in boxes. A first reading could omit all the frames while more advanced understanding should be obtained by their later perusal.

2.2 Adhesionless Contact: Stiffness

In this section we assume no interaction between the surfaces and investigate elastic contact. We bring the surfaces from “far away” (Fig. 2.1a) into contact (Fig. 2.1b). Without loss of generality (see frame 1) we assume that it is the tip which is elastic. The reduced modulus is $E^* = E/(1 - \nu^2)$, where E is Young’s modulus and ν the Poisson ratio. The flat is rigid.

Frame 1: Contact—response and boundary conditions

The results presented here are exact under a number of hypotheses, including absence of friction and small deformations. However, they are quite robust. A good example of deviations with large deformation and the resulting breakdown of the sphere/flat symmetry can be found in [1].

Within linear elasticity, if both surfaces are curved, the curvatures R_i^{-1} add up to provide the overall curvature R^{-1} . If both tip and substrate feature significant compliances, the compliances add up as

$$E^{*-1} = \left(E_t / (1 - \nu_t^2) \right)^{-1} + \left(E_s / (1 - \nu_s^2) \right)^{-1} \quad (2.2)$$

Moduli are in the 100 GPa range for stiff materials, but can drop considerably for polymers, down to 2 GPa for vitreous polymers or 10 MPa for elastomers. Contact radii in the present context are of the order of 10 nm.

2.2.1 Hertz: Contact Radius and Loading

We consider quasi-static response at contact. Due to the elastic deformation of the sphere, contact develops when increasing the *load* F . Actually, by ‘contact’ we mean: inside the contact zone the normal displacement at the surface is specified so as to cancel the initial shape $f(r)$. In this way the *contact zone* is the area in which the normal surface displacement (imposed by the contact boundary conditions) are prescribed for the elastic problem. The short-range repulsion has actually been turned into displacement boundary conditions [2].

In addition to the load F , another characteristic of the contact is the *penetration* δ . It is the rigid body displacement incurred by the undeformed parts of the sphere, far away from the contact. This rigid body displacement is made possible by the local deformation close to the contact area. Note that positive δ means penetration while positive F means compression. For adhesionless contact the geometry is such that if the sphere were to rigidly interpenetrate the flat by the same penetration $\delta_H(a)$, then the undeformed sphere would intersect the r axis at a radius equal to $\sqrt{2}a$ (Fig. 2.1b). From force and penetration, we can calculate the work expended by the *remote loading* to form the contact (frame 2), and also the work recovered when the contact breaks. A difference between these two means hysteresis and dissipation.

Coming closer to the contact itself, the geometry of the contact zone is defined by the *contact radius* a . To accommodate the deformation and especially the flattening of the parabolic profile (Eq. 2.1) of the elastic sphere inside the contact zone, a distribution of normal surface stresses arises at the interface. Hertz [3] demonstrated that the solution takes the form of an ellipsoidal distribution of *contact stresses*

$$\frac{\sigma(r)}{p_m} = \frac{3}{2} \sqrt{1 - (r/a)^2} \quad (2.3)$$

where p_m is the mean contact pressure.

The force as a function of contact radius is found from the integration of the contact stress distribution Eq. 2.3: in the Hertzian theory the (compressive) contact force is

$$F_H(a) = \frac{4E^*a^3}{3R} \quad (2.4)$$

and the mean pressure is

$$p_m \equiv \frac{F_H(a)}{\pi a^2} = \frac{4E^*}{3\pi} \frac{a}{R} \quad (2.5)$$

Finally the relation between penetration and contact radius results from the condition of zero stress at the contact edge and is

$$\delta_H(a) = \frac{a^2}{R} \quad (2.6)$$

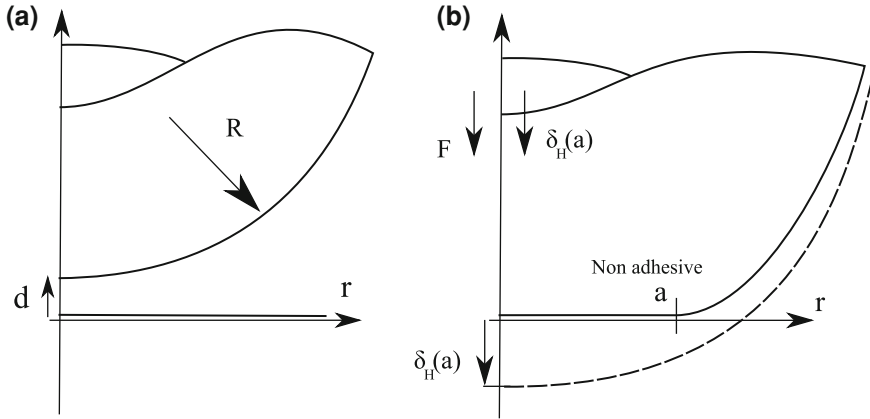


Fig. 2.1 Adhesionless contact: **a** sphere of radius R approaching a plane at a distance δ ; **b** after contact, when there is no adhesion: the contact radius is a and the penetration is δ_H . The undeformed sphere shape (dashed line) intersects the r axis at $\sqrt{2}a$

Appearances to the contrary this relation is highly non trivial and results from the calculation of the deformation *outside* the contact zone due to the stress distribution Eq. 2.3 inside.

Equations 2.4 and 2.6 are the contact equations for the adhesionless contact of an elastic sphere on a rigid plane as a function of contact radius a .

Frame 2: Hertz model—Approximate derivation

If we observe the deformation of the elastic body, the surface displacements are of the order of the penetration δ . By a very approximate geometrical argument, we estimate

$$\delta R = a^2 \quad (2.7)$$

By Saint-Venant's principle, we know that these displacements penetrate into the body over a typical distance equal to the contact radius a . As a result, the typical deformation is δ/a and the elastic energy for the penetration δ can be calculated as:

$$\varepsilon = \frac{1}{2} E \left(\frac{\delta}{a} \right)^2 a^3 \quad (2.8)$$

from which the relation for the force Eq. 2.4 results:

$$F(a) = \frac{d\varepsilon}{d\delta} \simeq \frac{E^*}{R} a^3 \quad (2.9)$$

and the stiffness defined by Eq. 2.11

$$S(a) \simeq E^* a \quad (2.10)$$

This last result is much more general than the specific case of sphere contact. Indeed it applies for all axisymmetric geometries (flat punch, cone) and depends only on the contact radius. The reason for this invariance is the absence of adhesion. The variation of the *contact radius* with penetration does depend on shape, but does not contribute to the stiffness because the stresses at the edge of the contact are zero. We will see in a later section that this result is significantly modified with adhesion, in which case these stresses are finite.

2.2.2 Contact Stiffness

For dynamic problems such as those of interest in this book, the quantity which is directly relevant is the contact stiffness defined by

$$S(a) \equiv \frac{dF}{d\delta} \quad (2.11)$$

In the Hertzian theory, the contact stiffness is given by the equation:

$$S(a) = 2aE^* \quad (2.12)$$

The most interesting feature is that the stiffness depends only on the contact radius a . This means that for different punch shapes and different loads, the same stiffness will be obtained if the contact radius is the same.

Since the contact radius a varies with load as Eq. 2.4, it is clear that the sphere contact—unlike a simple spring—does not have a constant stiffness, but that *stiffness increases as the load increases*. The consequences of this intrinsic nonlinearity will be emphasized in Sect. 2.8.1.

This result is remarkable because it was obtained in the framework of *linear* elasticity. The intimate reason is that the area over which the contact boundary conditions apply (specified by the contact radius a) change as a function of loading. In this sense, contact is a typical example of geometrical nonlinearity.

2.3 Interactions: Adhesion

Adhesive interactions will modify this picture significantly. Let us consider again the case where the two surfaces face each other at some distance, as in Fig. 2.1a. We will briefly discuss these interactions, the force they produce on the tip and finally their impact when they couple with elastic deformation.

2.3.1 Interactions: Derjaguin Approximation

When surfaces are brought together within some distance d , they start to experience “long-range” interactions. The nature of the interactions involved is best investigated from the interaction *force* they produce, which can be measured by AFM or other devices such as Surface Forces Apparatus (SFA) [4]. Here, we consider the case where this interaction is attractive, eventually leading to adhesion.

We can quantify this attraction by an interaction potential $V(h)$. V is defined for two unit surface areas facing each other with a gap h . The reference state is for infinite separation so that $V(\infty) = 0$. In fact infinity is reached rapidly since the range of the interactions δ_{int} is of the order of a few 10 nm or less. For a curved surface facing a flat at a distance d , there is a simple relation between the interaction force $F(d)$ and the interaction potential V . This relation, called the Derjaguin approximation [5] (frame 3), which neglects all deformation induced by the interaction stresses, states that

$$F(d) = 2\pi R V(d) \quad (2.13)$$

Note that V must be negative for the attractive interactions to result in an attractive (*i.e.* negative) force.

Frame 3: Derjaguin approximation

Given the interaction potential, the normal surface stress distribution is obtained by Eq. 2.18. The surface integral of this stress distribution gives the total force

$$F(d) = 2\pi \int_0^\infty dr r \sigma(r) \quad (2.14)$$

Taking into account the parabolic shape Eq. 2.1, the surface integral can be turned into an integral over the gap h resulting into

$$F(d) = -2\pi \int_d^\infty R dh(r) \frac{dV}{dh} \quad (2.15)$$

from which Eq. 2.13 results.

2.3.2 Nature of the Interactions

A large variety of interactions has been identified, collectively known as surface forces. A full gamut of such interactions is to be found in polar liquids, especially

water, but this is less relevant to the present topic. In vacuum, electrostatic interactions give rise to complex problems due to their long-range nature, whereby simple calculations such as frame 3 do not apply.

2.3.2.1 Van der Waals Interaction

Van der Waals interactions are often quoted as the typical surface interactions. It is true that due to material polarisability, van der Waals interactions are always present. Moreover, they lend themselves to a degree of mathematical sophistication verging on fine art [6]. Finally, at longer distances they are well approximated by the simple analytic form:

$$V(h) = -\frac{A}{12\pi h^2} \quad (2.16)$$

where A is the *Hamaker constant*, of the order of 1×10^{-20} J. For all these reasons, van der Waals forces have become the archetype of surface forces.

Inserting Eq. 2.16 into Eq. 2.13 we obtain the van der Waals force between tip and substrate,

$$F(d) = -\frac{AR}{6d^2} \quad (2.17)$$

an expression which is often used in the literature (see Sect. 2.3.4).

We now turn to the *interaction stresses* σ_0 . The normal stress at the surface resulting from the interactions is given by the derivative of the interaction potential:

$$\sigma(h) = -\frac{dV}{dh} \quad (2.18)$$

Typical values for interaction stresses resulting from van der Waals forces can be calculated using Eq. 2.18 with a cutoff distance of about 0.1 nm. A stress in the range of $\sigma_0 \simeq 1$ GPa appears.

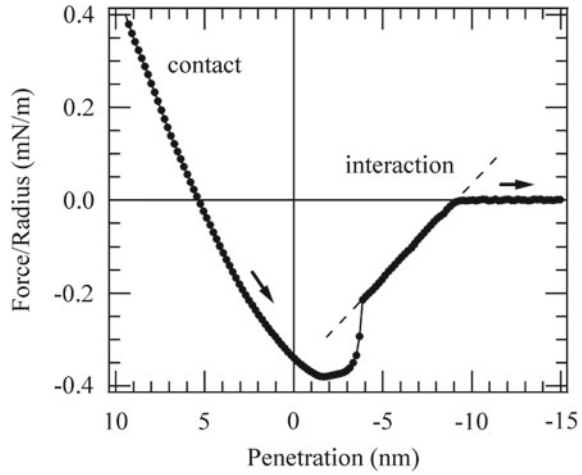
2.3.2.2 Liquid Meniscus

In ambient atmosphere, for hydrophilic surfaces, the interaction will be primarily mediated by a thin layer of adsorbed water, which forms a capillary bridge between the surfaces. This example has also been studied in great detail because it is both very frequent and relatively simple [7].

In this case the interaction stresses are a constant σ_0 which is given by the hydrostatic pressure inside the liquid meniscus. If the liquid in the meniscus is at equilibrium, the chemical potential in the liquid is constant and so is the pressure. Then

$$\sigma_0 = \gamma/r_0 \quad (2.19)$$

Fig. 2.2 Adhesive contact between sphere–plane silica surfaces. The interaction is mediated by a meniscus of liquid, with a linear force–distance plot typical for an equilibrium state of the meniscus. After [8]



where the surface tension of the liquid γ lies around 0.1 J/m^2 while the radius of curvature r_0 of the liquid meniscus is, for ordinary vapor pressures, in the nanometer range. As a result the order of magnitude of the interaction stresses is significantly smaller for liquid meniscus than for van der Waals interactions, about 100 MPa at most.

In this case Eqs. 2.13 and 2.18 show that the interaction potential is linear, so that the force is:

$$F(d) = -2\pi R 2\gamma \left(1 - \frac{d}{2r_0}\right) \quad (2.20)$$

for $0 < d < 2r_0$. The radius of curvature of the meniscus r_0 is therefore also the range of the adhesive interactions. This linear behavior is clearly evidenced in some SFA experiments (Fig. 2.2).

2.3.3 Adhesive Contact with Weak Interactions

We now bring the surfaces into contact in the presence of adhesive interactions. The logical extension of the Derjaguin approximation is the Derjaguin Muller Toporov (DMT) model [9], which assumes that contact occurs exactly as with the Hertzian model (Sect. 2.2.1). Here, it is considered that the interaction stresses do not bring about significant deformation of the elastic bodies, and the force resulting from this interaction F_{stress} can be calculated as if acting on a body deformed by the contact stress distribution only. Put otherwise, we assume that the magnitude of the contact stresses largely exceed the interaction stresses. If calculated strictly, the details of this model are rather tedious [10] but a good approximation has been provided by Maugis [11]. He has suggested a Hertzian model plus a constant force offset

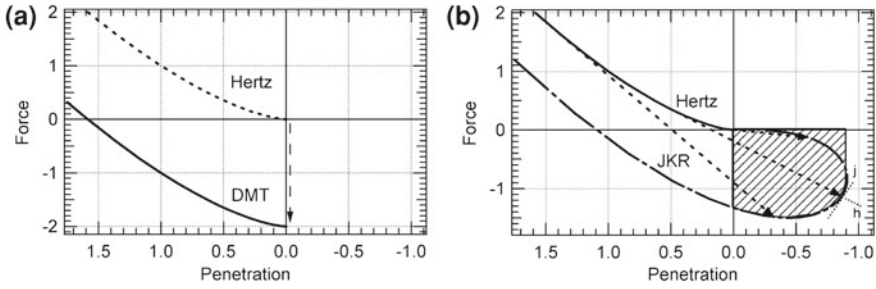


Fig. 2.3 **a** Hertz versus DMT model in normalized units. The DMT model results from a simple force offset. **b** Hertz versus JKR model. The JKR is obtained by point by point translation along the tangent to the Hertz model by a displacement equal to the neck height δ_{adh} . The shaded area is the energy expended in the stretching of the neck. Also shown as *small dashed lines* are the contact stiffnesses for fixed contact radius (Hertzian stiffness, marked 'h') and for free contact radius (JKR stiffness, marked 'j') (see Sect. 2.8.5). Note that in the extreme case selected here, these two stiffnesses actually have opposite signs

$$F_{\text{stress}} = -2\pi w R \quad (2.21)$$

resulting in a remote loading (Fig. 2.3a)

$$F(a) = F_H(a) - 2\pi w R \quad (2.22)$$

Continuity with Eq. 2.13 is ensured because the adhesion energy $w > 0$ obeys

$$w = -V(d = 0) \quad (2.23)$$

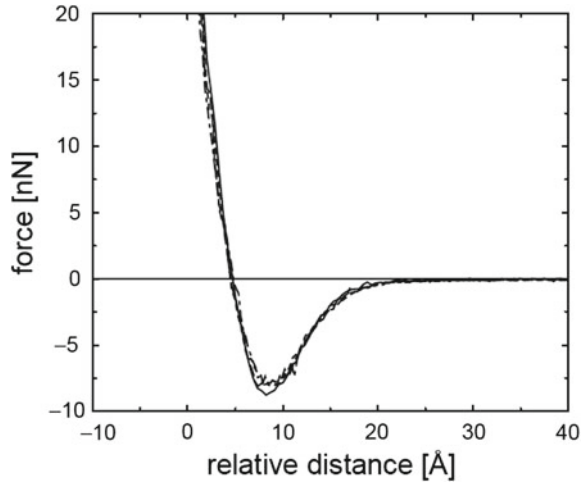
From Eq. 2.22, it becomes clear that the main effect of adhesion is to increase the contact radius for a same external load, since the load acting on the contact $F_H(a)$ is the remote loading $F(a)$ plus the adhesive contribution $2\pi w R$. Energy is gained from the adhesive interactions but balanced by the increased elastic energy stored due to larger contact area.

In the DMT model however, the contact stiffness is not affected by the interaction. For the same contact radius it is still given by the Hertzian expression Eq. 2.12.

2.3.4 Impact of Adhesion on Dynamic Response in AFM

Direct evidence for such long-range interactions has been found in various types of AFM measurements. Here, we illustrate the concept in an experimental configuration where oscillation amplitudes much larger than the interaction range are used. The oscillatory motion of the tip can be fully reconstructed, duly taking into account the small part of the trajectory where interaction of the tip with the surface occurs [12].

Fig. 2.4 Reconstruction of the interaction potential from the large amplitude oscillatory response of an AFM tip-cantilever. Beyond 10 Å a van der Waals attractive potential is evidenced while the contact compliance sets in at smaller relative distances. From [12]. Copyright (1999) by the American Physical Society



Long-range interactions and contact have been taken into account using Eqs. 2.17 and 2.22 respectively. Profiles of such interactions between a tungsten tip and a silicon substrate has being inferred from the measured frequency and phase shifts (Fig. 2.4) and found to agree well with van der Waals forces.

Frame 4: Adhesive contact—Approximate derivation

The true nature of the adhesive contact of a curved body is a competition between a gain in adhesion and the ensuing elastic energy penalty (Hertzian term). In the DMT model, the energy gain is obtained from the integral of the interaction potential over the gap as in the calculation of the Derjaguin approximation (frame 3).

In another approach [5], the adhesive energy gain is estimated from the adhesion energy w and the contact area so that the total energy is

$$\varepsilon \simeq \frac{1}{2} E \left(\frac{\delta}{a} \right)^2 a^3 - \pi w a^2 \quad (2.24)$$

The relation for the force is

$$F(a) = \frac{d\varepsilon}{d\delta} \simeq \frac{E^*}{R} a^3 - \pi R w \quad (2.25)$$

showing that the adhesive contribution is of the order of $\pi R w$.

Although this rough estimate is useful for a preliminary discussion, the derivation we have used is actually flawed. In writing the adhesive term $\pi w a^2$ we implicitly assumed that for an infinitesimal variation of the contact radius a , the variation in adhesion energy is proportional to the variation of contact area. This implies that the gap shape around the contact edge is sharp enough to exhaust the interaction range. However such a sharp gap shape has an elastic energy penalty which must be taken into account as in Sect. 2.4.

2.4 Coupling with Strong Interactions

So far, the coupling was quite simple since elastic deformation results from the contact stresses only (Eq. 2.3). Further difficulties arise when the adhesive stresses themselves are large enough to induce significant deformation of the surface, bringing more than a simple additional load (frame 4).

2.4.1 JKR Model

Taking this additional surface deformation into account is a more complex problem. Fortunately, the limit case where considerable deformation occurs can be treated relatively simply.

Frame 5: The JKR model

The flat punch elastic energy is

$$\varepsilon = \frac{1}{2} E \left(\frac{\delta}{a} \right)^2 a^3 \quad (2.26)$$

so that the energy release rate is

$$\mathcal{G} = \frac{1}{2\pi a} \frac{d\varepsilon}{da} \simeq E^* \frac{\delta^2}{a} \quad (2.27)$$

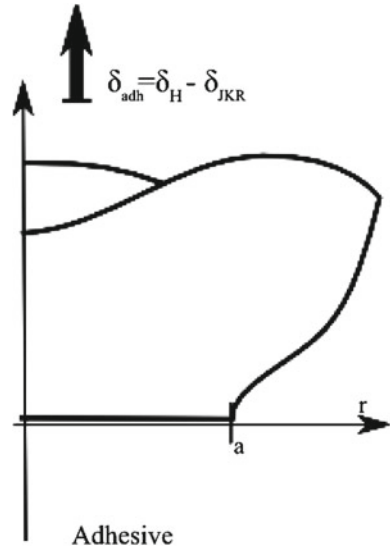
Equilibrium results from

$$\mathcal{G} = w \quad (2.28)$$

so that

$$\delta_{\text{adh}} \simeq \sqrt{\frac{aw}{E^*}} \quad (2.29)$$

Fig. 2.5 Adhesive contact: cusp at the contact edge in the JKR model, demonstrating the typical JKR flat punch displacement



Johnson, Kendall and Roberts (JKR) have shown that the deformation induced by the interactions amounts to an additional flat punch deformation (Fig. 2.5). The resulting flat punch displacement (see frame 5) is central to the JKR theory [13]:

$$\delta_{adh} = \sqrt{\frac{2\pi aw}{E^*}} \quad (2.30)$$

Here, we take δ_{adh} positive but in fact adhesion induces a reduction of the penetration (for a given contact radius) so that a minus sign appears in the contact equations:

$$\delta_{JKR}(a) = \delta_H(a) - \delta_{adh}(a) \quad (2.31)$$

$$F_{JKR}(a) = F_H(a) - S(a)\delta_{adh}(a) \quad (2.32)$$

The force has been derived using the flat punch displacement and stiffness according to Eq. 2.12. This adhesive contribution in Eqs. 2.31 and 2.32 amounts to a translation along the tangent to the Hertzian curve, which is schematized in Fig. 2.3b. The set of Eqs. 2.30, 2.31 and 2.32 together forms the JKR theory. Note that these equations are often presented spelled out, which may be less illuminating.

2.4.2 Pull-Out Force

The *pull-out force* is the maximum tensile force which needs to be applied to break the adhesive contact and rip the sphere off the surface. Somewhat by accident, and

for the sphere only, the pull-out force is nearly independent of the type of adhesive contact model

$$F = -\alpha\pi wR \quad (2.33)$$

It is clear that $\alpha = 2$ in the DMT limit. From Eq. 2.32 and looking for the minimum, we can calculate that $\alpha = 3/2$ in the JKR case. This value results from the balance of the two energy terms: around pull off, both (compressive) contact load and (tensile) interaction load are of the order of πwR .

Frame 6: Is adhesion relevant ?

Equation 2.33 is specific to the sphere geometry. Most if not all other cases (tip shapes or symmetry) do not offer the same simplicity. Under the assumption of sphere geometry, a question in order is: under which type of loading is adhesion relevant? To answer this question we balance interaction load (of the order of πwR) and contact load. From 2.24 we deduce that adhesion steps in when

$$a \simeq \left(\frac{\pi R^2 w}{E^*} \right)^{1/3} \quad (2.34)$$

The contact load turns out to be dominant above these values of contact radius. In our case, for a comparatively rigid solid, this contact radius is of the order of 1 nm and the load is of the order of 10 nN.

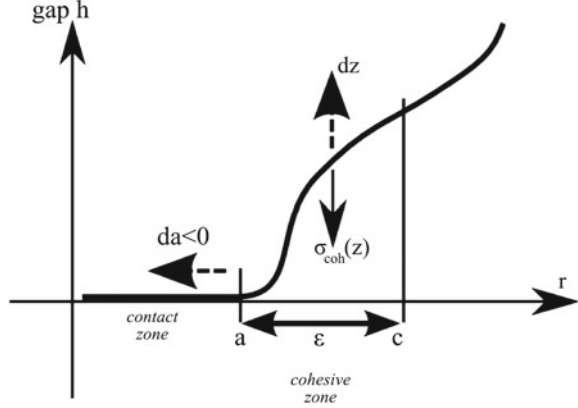
If the pull-out force is barely dependent upon the model, a question may arise: is the choice of contact model of limited consequences and somewhat arbitrary or is there a good reason to pay attention to which contact model to use? If the pull-out force is not very revealing itself, these contact models involve *very different stress distributions*: beyond the mere pull-out force, model-dependent responses are to be expected. This is the case for example in Sects. 2.7 and 2.8. For this reason a more general picture is needed and we now consider how the adhesive interactions are coupled with the contact problem in more detail.

2.5 AFM Tips: An Intermediate Case?

2.5.1 Adhesive Interactions Revisited: Contact Problem

In fact in both the DMT and the JKR models, the details of the interactions do not appear. We are dealing with limit cases and in the end the adhesion energy w remains as the only relevant parameter for the description of the physical process of adhesion. In the more general case, the adhesive interactions induce tensile stresses over some area around the contact zone: this area is called the *cohesive zone* (Fig. 2.6). Due to the finite range of the interactions the cohesive zone extends over a distance

Fig. 2.6 Adhesive contact: schematics of the impact of adhesive interactions on the local deformation around the contact zone (cohesive zone model). c is the outer limit of the region over which interaction stresses act. Finite range of the interactions result in a lateral extension $\epsilon = c - a$ of the area over which the interaction stresses operate



$\epsilon = c - a$. This distance will come out useful when the dynamics of the contact edge is calculated (Sect. 2.8).

In the general case, calculation of the impact of the interaction stresses on the deformation is not an easy task. For recent attempts see [14, 15]. None of these models lends itself to simple explanations however. Let us only mention that the calculation proceeds as in frame 3, but this time taking into account the deformation directly due to the interaction stresses. The contact equations are then

$$\delta_{\text{JKR}}(a) = \delta_H(a) - \delta_{\text{adh}}(a) \quad (2.35)$$

$$F_{\text{JKR}}(a) = F_H(a) - S(a)\delta_{\text{adh}}(a) - F_{\text{stress}} \quad (2.36)$$

revealing the mixed JKR–DMT character of the solution. However, neither $\delta_{\text{adh}}(a)$ nor F_{stress} is given by the JKR (Eq. 2.30) or DMT (Eq. 2.21) model but rather by one of the more general expressions available in the literature, such as [2, 11, 16].

In these models the interaction stresses are defined by Eq. 2.18 where h is the gap between the surfaces. They form the boundary conditions outside the contact area. There is a difficulty: due to elastic response the gap itself is affected by the interaction stresses. As a result a self-consistent treatment is called for. It has been shown that the finer details of the interaction potential (or the surface stress distribution) are not relevant and play a role only to higher order [2]. Therefore, in most cases, for the coupling between the interactions and the contact mechanics, only two entities must be considered: adhesion energy w and the interaction stresses with magnitude σ_0 . In this context, following Eqs. 2.18 and 2.23, it appears that the range of the interactions δ_{int} obeys

$$\delta_{\text{int}}\sigma_0 \simeq w \quad (2.37)$$

For numerical simplicity, the interaction stresses are often considered constant throughout the cohesive zone [11]. This is the so-called Dugdale-Barrenblat model. In the case of the Dugdale-Barrenblat model, in Eq. 2.37 equality applies.

2.5.2 Which Model: Does Adhesion Induce Deformation?

We now discuss the general features of the adhesive contact depending upon interaction, loading, and geometry, in the spirit of the “adhesive map” by Greenwood and Johnson [17]. The relevance of the adhesive contribution is examined in frame 6. Here, we consider whether adhesion affects deformation or not, i.e., whether we are close to the DMT or to the JKR model or in some intermediate case. Based on our earlier considerations on interaction stresses, a characteristic parameter emerges when comparing interaction stresses and contact stresses. We introduce the Tabor parameter [18]

$$\lambda \simeq \frac{\sigma_0}{\sigma} \quad (2.38)$$

where σ stands for the contact stresses.

In the absence of adhesion $\sigma = p_m$ (Sect. 2.2.1). In the range where adhesion is significant (frame 6), the load F is of the order of $\pi w R$ and the contact stresses are

$$\sigma = \frac{F}{\pi a^2} \simeq \left(\frac{w E^{\star 2}}{\pi^2 R} \right)^{1/3} \quad (2.39)$$

An interesting consideration arises if the Tabor parameter λ is expressed in terms of penetration: then it is found that

$$\lambda \simeq \frac{\delta_{\text{adh}}}{\delta_{\text{int}}} \quad (2.40)$$

If the flat punch displacement δ_{adh} is large compared to the range of the interactions δ_{int} , then the cohesive zone size is small, and adhesion energy is transferred between the interface and the tip by large elastic deformations located close to the contact edge as embodied by the flat punch displacement, resulting in the neck at the contact edge (Fig. 2.5). This is a fracture-like process, central to the JKR limit.

Note however that the neck has to be stretched out upon rupture. However when the sphere comes to the surface, contact forms at a penetration equal to zero since the interaction is short ranged. As a result, hysteresis appears, as illustrated by the shaded area in Fig. 2.5b.

On the contrary, if the flat punch displacement is comparatively small ($\lambda \ll 1$), adhesion energy transfer operates directly through the work done by the interaction stresses in the displacement of the sphere surface. This is the DMT limit where the cohesive zone size is large. Since the interaction range is large the contribution of the interaction to the stiffness is zero, and the stiffness is the Hertzian stiffness as already mentioned.

Intermediate cases appear for $\lambda \simeq 1$.

2.5.3 Small Tips

Given the typical values for interactions and the typical contact radii of AFM tips, we find that for comparatively rigid surfaces δ_{adh} is of the order of 0.1 nm. It is quite clear that the pure JKR theory is unlikely to apply in our case.

This means that the tip has to be considered as comparatively rigid compared to the attractive interactions. The deformation incurred during the adhesive contact is primarily the deformation of the Hertzian adhesionless contact but for the stiffer types of adhesive interactions, with small decay lengths, λ may range around 1 and the adhesive contact acquires partial JKR character. This is typically the case for ultra-high vacuum measurements. An example of such a case is shown in Fig. 2.7. In contrast, under ambient conditions a longer ranged interaction dominates, which is due to the presence of adsorbed water. The resulting meniscus induces an interaction shown in Fig. 2.2 with a range of several nanometers, and the contact will be in a typical DMT state. One of the rare cases where a contact close to a true JKR case could be obtained is polymeric surfaces where low modulus and high effective adhesion energies are expected to result in large λ through Eqs. 2.30 and 2.40.

For small tips, in the intermediate range, the cohesive zone size is of the order of the adhesive contact radius and the typical contact stresses upon pull-off are in the range of σ_0 . This is similar to fiber problems: the average stress at the surface of the tip, or rupture stress increases dramatically and eventually reaches the theoretical interface stress when the size of the contact area decreases [19, 20].

2.6 Films

Thin films and coatings are ubiquitous in technological applications. Here, we consider the case of contact to coated substrates.

2.6.1 Stiffness

As mentioned earlier (frame 2), the deformation field affects the elastic body to a typical depth of the order of the contact radius a . It is important to note that this depth is only indirectly related to the penetration δ , through Eq. 2.6 for example. In the case of an elastic property mismatch between the coating and the substrate, the macroscopic response will be affected by the presence of the substrate beneath the film if the film thickness is less than several times the contact radius. To estimate the impact of the film, one can suggest to use Eq. 2.6 with film values to infer the contact radius. It will be necessary to consider the full solution if this contact radius is not significantly smaller than the film thickness. If this is the case, exact solutions have been calculated [23–25] which can be used fairly easily.

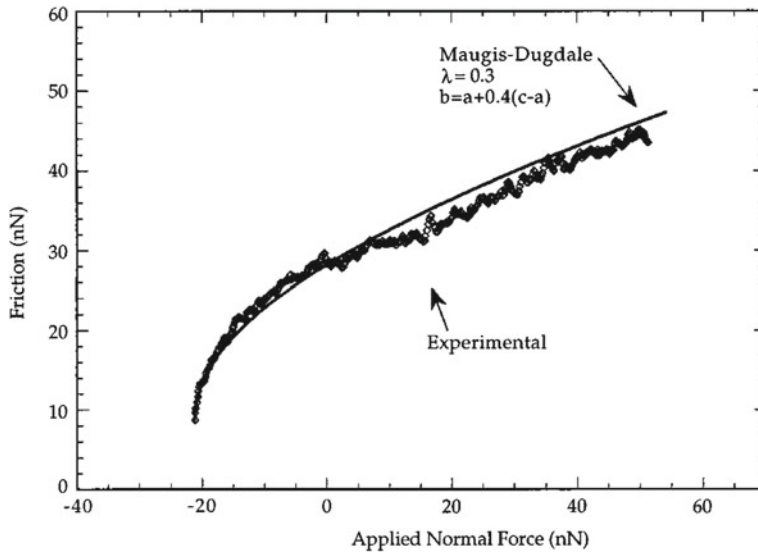


Fig. 2.7 Friction force (assumed to be proportional to contact area) and model for the adhesive contact area as a function of load. The data are best fitted with an intermediate adhesive contact model where neither pure JKR nor pure DMT models apply. From [21]. Copyright (1997) by the American Physical Society

Note that the problem is especially significant in the case of an elastomeric film, which is somewhat liquid-like and therefore incompressible. Due to the suppression of shear deformation because of confinement, the effective response of the film (Fig. 2.8) is driven by the bulk modulus which is considerably larger than the shear modulus because of incompressibility.

2.6.2 Adhesion

For adhesive contacts, the film may have two very different types of impacts. On the one hand, the film is likely to change the interactions between the tip and the surface, and thus the values of the adhesion energy w and the interaction stresses σ_0 . On the other if the film thickness is not large enough according to the criterion outlined above, then the substrate effect will affect the contact response. The contact equations should then take adhesion into account [26]. In the more elaborate case where contact zone radius, film thickness, and cohesive zone size become comparable, recent calculations could become useful although they are by no means numerically simple [27].

Note however that a most salient feature in this case is that, for identical adhesion energy w , the pull-out force is barely affected by the presence of the coating. This idea

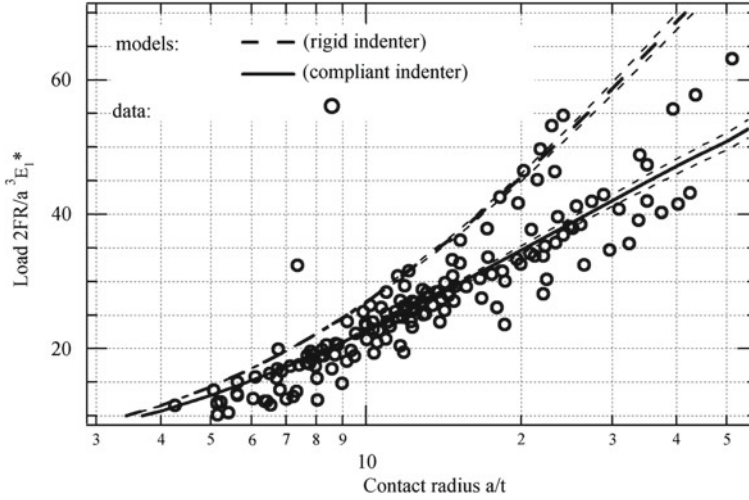


Fig. 2.8 Load as a function of normalized contact radius for sphere–plane contacts on acrylate films. Confinement results in an increase of the film modulus so that the glass sphere compliance must be taken into account. Adapted from [22] with permission from Springer

must be brought in relation to our previous remark in Sect. 2.4.2: the pull-out force results from a balance between contact and adhesion energies. The result turns out to be independent from the mechanical properties of the half-space if it is homogeneous. In fact, this dependence is only barely reintroduced if homogeneity is lost as when a coating is present.

2.7 Roughness

Generally speaking, surface roughness impacts contact problems strongly. However, the real complexity arises from the statistical nature of the roughness coupled to the nonlinear nature of even the most basic contact, namely Hertzian contact [28]. In this sense roughness is likely to alter the qualitative response of a contact. The simplest possible example is an exponential distribution $n \propto \exp(-z/\tau)$ of summit heights with identical curvatures R^{-1} . Here, τ is the standard deviation of the roughness. Then the density d_c of summits in contact with a flat surface obeys

$$d_c \propto \exp(-d/\tau) \quad (2.41)$$

where d is counted from the mid-plane of summit heights. Summing individual summit areas and forces over this exponential distribution we obtain that both total area and total force are also proportional to d_c . As a result, and in contrast to the

results of Sect. 2.2.1, force and true contact area are now proportional, an idea which is central to our understanding of the laws of friction [29].

In addition, roughness will compete with attractive interactions. It is this issue for which more insight can be provided. In terms of adhesive contact, it is well known in practice that for rather rigid materials, a very limited amount of roughness suppresses the adhesion of a sphere brought to a surface. For weak interaction stresses as in the DMT contact model, the typical lengthscale involved is the interaction range δ_{int} . It is clear that for a roughness τ small compared to this interaction range the attractive interactions can still be accounted for as in Sect. 2.3.3. In the other limit where the deformation incurred through attractive interactions are sizeable, the characteristic distance which emerges is the JKR flat punch displacement δ_{adh} . This idea has been elegantly demonstrated by Fuller and Tabor [30]. A handwaving argument goes as follows: the (tensile) contribution of each asperity to the adhesion force is a constant of the order of $\pi w R$; on the other end, the (compressive) contact force generated by each asperity grows faster than linearly with penetration δ , following Eq. 2.4. For a roughness distribution with standard deviation τ , the sum of the repulsive contributions will far exceed the attractive contributions if the roughness distribution obeys

$$\tau \gtrsim \delta_{\text{adh}} \quad (2.42)$$

For the case of interest here, it is possible that the tip will interact with a limited number of asperities. In this case the statistical approaches are of a somewhat limited relevance and one must rely on the more demanding and less general calculation of distributions of local configurations [31].

2.8 Dynamics

We now consider a tip impinging on a surface. In keeping with the rest of this chapter, the viewpoint is the mechanics of a sphere touching a surface. The sphere is considered as free, with initial velocity v_0 , and the rest of the system, and especially the cantilever and its mechanics, is not taken into account. Our aim is to understand dissipation during contact. However, this part of the question is much less advanced than the quasistatic part.

In agreement with the views developed here, friction is not considered, although in some cases it could account for a significant part of dissipation during dynamic contacts. In this restricted frame, the dissipated energy is all the energy which is not fed back to the remote loading when the surfaces have ceased to interact. There are many processes active in this area. For the adhesionless contact, two processes fail to restore all the energy injected in the contact: (1) acoustic emission and (2) material dissipation, which may occur through delayed elastic response (viscoelasticity) or non-elastic response (plastic deformation). If adhesion is present, several additional processes must be mentioned: (3) the physics of adhesion may be partly irreversible (the adhesion energy is different for a growing and a receding interface); and if

adhesion induces additional deformation, as in cases close to the JKR limit: (4) the rupture may occur by instability and part of the energy involved is not restored to the remote loading; (5) material dissipation (as in 2) may result from deformations specific to the adhesive process.

In this area, the number of in-depth studies is quite restricted. Here, we will only hint at a few directions. We will start from the (reversible) elastic rebound of an adhesionless sphere. Then we will qualitatively consider the impact of dissipative mechanisms (1), (2), (4), and (5).

2.8.1 Sphere Impact

If we assume a free sphere of mass m impinging on an elastic adhesionless surface, contact leads to rebound. Taking into account the non-constant stiffness at contact (Sect. 2.2.2), conservation of energy during rebound implies

$$\frac{1}{2}m \left(\frac{d\delta}{dt} \right)^2 + \frac{8}{15}E^*\sqrt{R}\delta^{5/2} = \frac{1}{2}mv_0^2 \quad (2.43)$$

where v_0 is the sphere velocity at impact. Maximum penetration occurs when $\frac{d\delta}{dt} = 0$ so that

$$\delta_{\max} = \left(\frac{mv_0^2}{\frac{16}{15}E^*\sqrt{R}} \right)^{\frac{2}{5}} \quad (2.44)$$

$$a_{\max} = \left(\frac{mv_0^2 R^2}{\frac{16}{15}E^*} \right)^{\frac{1}{5}} \quad (2.45)$$

To evaluate the rebound, $d\delta/dt$ can be integrated numerically from Eq. 2.43. In fact a linear approximation

$$\frac{\delta}{\delta_{\max}} = \sin \left(\pi \frac{t}{T_c} \right) \quad (2.46)$$

has been shown to perform well [32], where the typical contact time is

$$T_c = \alpha \frac{\delta_{\max}}{v_0} \simeq 3 \left(\frac{m^2}{E^{*2} R v_0} \right)^{\frac{1}{5}} \quad (2.47)$$

2.8.2 Inertial Effects

Dealing with acoustic frequency excitations, it is useful to estimate whether inertial effects are significant, that is to say whether acoustic waves will be generated during such a contact. An estimate can be obtained as follows: an acoustic mode obeys

$$\rho \frac{dv}{dt} = -\text{div} \sigma \quad (2.48)$$

where v is the velocity field, ρ the density, and σ the stress field. Order of magnitude estimates from frame 2 show that we remain in the quasistatic limit as long as

$$\rho \frac{\delta}{T_c^2} \ll E \frac{\delta}{a^2} \quad (2.49)$$

or

$$\frac{a_{\max}}{T_c} \ll \sqrt{E/\rho} \quad (2.50)$$

which is the sound velocity. For a free sphere with incident velocity v_0 and mass m , the criterion is

$$\left(\frac{E^* v_0^3 R^3}{m} \right)^{\frac{1}{5}} \ll \sqrt{E/\rho} \quad (2.51)$$

which is consistent with standard estimates of the dissipation induced by acoustic waves during contacts (Eqs. 11 and 12 in [32]). For a free sphere of mass m , the criterion becomes independent upon radius since $m \propto \rho R^3$. For an incident velocity of about $1 \times 10^{-2} \text{ ms}^{-1}$, a high value for a tip touching a surface, it appears that acoustic emission is negligible.

2.8.3 Material Dissipation: Contact Area

To account for dissipation during contact, we couple contact zone deformation with out-of-phase material response. This case has been considered in a classical paper [33] with a viscous type of dissipation. The viscous constant η relates dissipative stress to deformation rate. If v_1 is the velocity of the sphere after contact (see frame 7)

$$\frac{v_1}{v_0} = 1 - g v_0^{1/5} \quad (2.52)$$

where

$$g \simeq \frac{\eta}{E^*} \left(\frac{E^* \sqrt{R}}{m} \right)^{\frac{2}{5}} \quad (2.53)$$

To relate the viscous parameter η with materials properties, it should be noted that the characteristic frequency is

$$\omega \simeq \frac{1}{T_c} \quad (2.54)$$

For elastomers the estimate seems consistent with the observed dissipation parameter $v/v_0 \simeq 0.65$ [34].

Frame 7: Contact—Dissipation

In the spirit of frame 2, we assume the elastic solution is perturbed by a first order dissipative term.

$$\sigma_{\text{visc}} \simeq \eta \dot{\epsilon} \quad (2.55)$$

where strain is

$$\epsilon \simeq \frac{\delta}{a} \quad (2.56)$$

and strain rate

$$\dot{\epsilon} \simeq \frac{\delta}{a T_c} \quad (2.57)$$

The elastic and the dissipative terms are in parallel. Then the energy dissipated during one contact is

$$\mathcal{E}_{\text{diss}} \simeq \eta \dot{\epsilon} \epsilon a^3 \simeq \eta \delta_{\text{max}} a_{\text{max}} v_0 \quad (2.58)$$

From energy balance

$$\frac{1}{2} m v_1^2 + \mathcal{E}_{\text{diss}} = \frac{1}{2} m v_0^2 \quad (2.59)$$

so that for small velocity variations

$$\frac{v_1}{v_0} \simeq \frac{\mathcal{E}_{\text{diss}}}{\frac{1}{2} m v_0^2} \quad (2.60)$$

from which Eq. 2.52 results.

2.8.4 Adhesion Hysteresis: Elastic Instability

If the contact is in a JKR type of limit, then the neck formed upon contact needs to extend up to δ_{adh} before rupture occurs. Using Eqs. 2.30 and 2.34 this stretching energy is about

$$\mathcal{E} \simeq \left(\frac{\pi^2 R^4 w^5}{E^2} \right)^{\frac{1}{3}} \quad (2.61)$$

This energy is spent by the remote loading to stretch the neck but is not gained upon coming-in since the interaction range $\delta_{\text{int}} \ll \delta_{\text{adh}}$ is small and the neck does not form during surface approach. In short, this is the amount of energy lost in one contact cycle. This elastic instability is a common mechanism for adhesive energy dissipation [35, 36]. Of course, if λ is of the order of one (i.e. we are not in a full JKR case) only a *fraction* of this energy will be dissipated by instability.

2.8.5 Material Dissipation: Contact Edges

In the same regime, dissipation induced by material response may occur due to high rate deformation close to the contact edge. This is often the case for polymeric materials. The additional dissipation incurred can be phenomenologically modeled as an effective adhesion which depends upon contact edge velocity da/dt [37] as

$$G(da/dt) = w(1 + \phi(da/dt)) \quad (2.62)$$

where power laws are often used for the dissipative function ϕ . Relation between ϕ and the dynamic response of the polymer involved has been demonstrated experimentally [37] but theoretical justifications involving contact edge deformation processes are only partly successful to date [38–41].

Nonetheless, the phenomenological relation Eq. 2.62 is very useful. As an example we consider again the rebound dynamics for macroscopic balls [34]. During rebound, the characteristic velocity is

$$\frac{da}{dt} \simeq \frac{a_{\text{max}}}{T_c} \quad (2.63)$$

and the characteristic angular frequency is

$$\omega \simeq \frac{1}{\epsilon} \frac{da}{dt} \simeq \frac{a_{\text{max}}}{\epsilon} \frac{1}{T_c} \quad (2.64)$$

where ϵ is the cohesive zone size. Note that for a macroscopic sphere, this frequency is much higher than for contact zone dissipation (Eq. 2.54), due to the typical small size of the cohesive zone. The additional dissipated energy is

$$\mathcal{E}_{\text{diss}} \simeq \pi a_{\text{max}}^2 G(da/dt) \quad (2.65)$$

The results fit rebound experiments on elastomers extremely well [34].

Another remarkable result is that in an oscillatory experiment the stiffness of a JKR contact depends upon frequency in a non-trivial manner. Indeed, for dissipative materials, the high frequency motion of the contact edge may be hindered by large dissipation at high strain rates. Then the stiffness must be calculated *at constant contact radius*: it is the Hertzian stiffness. On the other hand, at comparatively low frequencies, the contact edge is free to move during oscillations and the contact stiffness must be calculated from Eq. 2.32 *at constant adhesion energy*. These two cases have been shown in Fig. 2.3b) as two straight lines marked h (Hertzian stiffness) and j (JKR stiffness). This transition has been very clearly observed for polydimethylsiloxane (PDMS) elastomers by oscillatory nanoindentation experiments with a micron-sized sphere [42].

2.9 Conclusion

We have outlined a few results on the adhesive contact of tips to surfaces. We have drawn on the body of theories devoted to spheres to discuss contact and the resulting contact stiffness.

We have emphasized the fact that in the presence of attractive interactions, leading to adhesion, the results are impacted in a non-trivial way. We have briefly discussed typical interactions which can be met with during AFM operation. We have shown in which way these interactions couple with elastic deformation. If the surface compliance is low enough, the adhesive interactions induce additional local deformation which alter the physics of the contact. Due to their small radius, AFM tips were shown to lie in the stiff to intermediate regime. Pure JKR case is not expected.

The impact is not directly perceptible from the bare pull-out force. Even when homogeneity breaks down, such as with a coating, the pull-out force is still only very moderately affected. More contrast appears when dealing with rough surfaces. However, it is when the dynamics of the response is considered that the strong impact of the low surface compliance really appears in full light, resulting in additional, specific dissipation mechanisms.

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