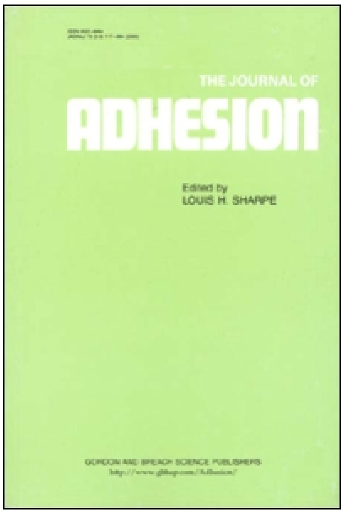

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The Jump-to-Contact Distance in Atomic Force Microscopy Measurement

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The jump-to-contact phenomenon of atomic force microscopy measurement is investigated. The force-approach relation for the adhesive contact based on the Lennard-Jones potential with the Derjaguin approximation is analyzed. For a small Tabor parameter, the force-approach relation is similar to that with the van der Waals *force between two* rigid *spheres. For a large Tabor parameter, the force-approach relation is similar to that with the* van der Waals *force between two* deformable *spheres. Empirical formulas for the approaching part of the force-approach curve are proposed. The jump-to-contact distance can be obtained by using the semi-empirical formulas. The jump-to-contact distance for a fixed grips device and for large Tabor parameter is also obtained.*

Keywords: AFM; Contact mechanics; Jump-to-contact; Numerical simulation

NOMENCLATURE

A	non-dimensional approach, $A = \frac{\alpha}{\varepsilon}$
A'	non-dimensional approach, $A' = \frac{A}{\mu^{3/7}}$
E^*	equivalent Young's modulus
E_1, E_2	Young's modulus
H	non-dimensional distance, $H = \frac{h}{\varepsilon}$
H'	non-dimensional distance, $H' = \frac{H}{\mu^{3/7}}$
h	distance between two surfaces
H_A	Hamaker constant
$K(\cdot)$	the complete elliptic integral of the first kind

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P	non-dimensional pressure, $P = \frac{p\varepsilon}{\Delta\gamma}$
p	pressure
R	equivalent radius of curvature, $\frac{1}{R} = \frac{1}{R_1} + \frac{1}{R_2}$
R_1, R_2	radius of curvature
r	radial coordinate
$\hat{r}$	non-dimensional radial coordinate, $\hat{r} = \frac{r}{\sqrt{\varepsilon R}}$
$\hat{r}'$	non-dimensional radial coordinate, $\hat{r}' = \frac{\hat{r}}{\mu^{3/14}}$
s	radial coordinate
$\hat{s}$	non-dimensional radial coordinate, $\hat{s} = \frac{s}{\sqrt{\varepsilon R}}$
$\hat{s}'$	non-dimensional radial coordinate, $\hat{s}' = \frac{\hat{s}}{\mu^{3/14}}$
W	non-dimensional force, $W = \frac{w}{2\pi\Delta\gamma R}$
W'	non-dimensional force, $W' = W\mu^{6/7}$
w	force between two spheres
α	distance between two spheres
$\Delta\gamma$	surface energy
ε	the intermolecular distance, where zero force occurs between two infinite surfaces.
μ	Tabor parameter, $\mu \equiv \left(\frac{R\Delta\gamma^2}{E^{*2}\varepsilon^3}\right)^{1/3}$

1. INTRODUCTION

Atomic force microscopy (AFM) is widely used in nanotechnology. In AFM measurements, jump-to-contact and jump-off-contact are well-known phenomena. For AFM measurements, the force-distance curves are fundamental tools in research [1]. Using the force-distance curves, Greenwood offered an explanation for jump-to-contact and jump-off-contact phenomena [2]. For an apparatus of finite stiffness, jump-to-contact and jump-off-contact occur when the force gradient exceeds the stiffness of the cantilever. For a fixed-grips device (apparatus of infinite stiffness), jump-to-contact and jump-off-contact occur at the vertical tangents of the S-shaped force-approach curve. Figure 1 shows the paths for jump-to-contact and jump-off-contact. A common technique in AFM work, introduced by Israelachvili and Tabor [3], is to measure sample topography in the non-contact mode, but this requires that jumping into contact is avoided.

The jump-to-contact phenomenon was first described by Overbeek and Sparnaay [4]. This phenomenon was also found by Tabor and Winterton [5] when measuring long-range van der Waals surface forces between crossed mica cylinders. Israelachvili and Tabor [3] gave a formula to predict the jump-to-contact distance, considering only the

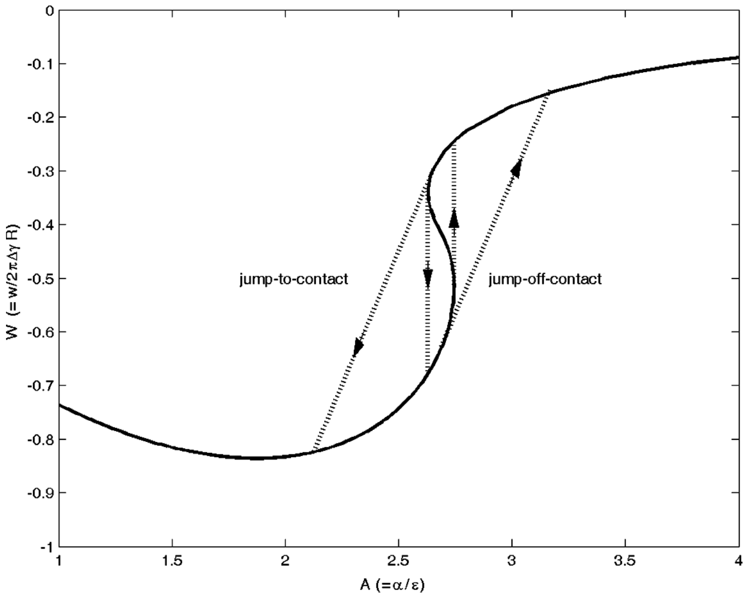

FIGURE 1 The paths for jump-to-contact and jump-off-contact. In an apparatus of finite stiffness, jump-to-contact and jump-off-contact occur when the force gradient exceeds the spring constant of the cantilever. In a fixed-grips device, jump-to-contact and jump-off-contact occur at the vertical tangents of the force-approach curve.

van der Waals force and assuming surfaces to be rigid. Their formula was widely used [1,6,7], for example, by Das *et al.* [8] in finding the Hamaker constant. In 1971, Johnson, Kendall, and Roberts proposed the famous JKR model [9], which predicts a pull-off force between two elastic spheres. The JKR model can be used to predict the jump-off-contact distance, but in terms of the surface energy instead of the surface force law. However, the JKR analysis does not deal with the jumping-to-contact problem. In 1992, Attard and Parker [10] assumed that the deformation varies slowly compared with the curvature, and gave an analytical formula for the jump criterion. Their jumping distance is the actual separation (not the approach) between two deformed spheres. It is not useful in predicting the jump-to-contact phenomenon. In 2004, Rutland *et al.* [11] analyzed the data for using AFM to measure deformable materials. In their analyses, they set the zero of separation at the end point of the jump.

However, Burnham *et al.* [12] found that electrostatic force plays an important role in long-range interaction. Gady *et al.* [13] found that van der Waals dominates the adhesion for a separation less than 30 nm, while the electrostatic force dominates the adhesion for separation larger than 30 nm. Since the electrostatic force depends on the surface charge density of the sample, Burnham *et al.* [12] concluded that if the cleanliness of the surfaces were better controlled, the van der Waals forces should become the dominant attraction between uncharged, non-magnetic surfaces. This paper is focused on the van der Waals force.

Self-consistent numerical analysis for the adhesive contact between elastic spheres was done by Greenwood [2] and Feng [14] to find the force-displacement relation with the Lennard-Jones force law, which combines the van der Waals long-range attractive forces with shorter range repulsive forces. Using this force-displacement relation, the jump-to-contact distance can be obtained. However, the interest of these workers was in the complete force-displacement curve, and particularly in the pull-off forces.

In this paper the jump-into-contact region will be examined in detail, and formulae for the approaching part of the force-approach curve will be derived: from these the jump-to-contact distance can be obtained.

2. ADHESIVE CONTACT BETWEEN SPHERES

2.1. Adhesive Contact of Rigid Surfaces

The Lennard-Jones (6,12) law, where the first term is the van der Waals attraction and the second term is an empirical one, describes the potential between two molecules. By integrating the Lennard-Jones potential over all molecules of two bodies, the pressure, $p(h)$, between two plane, semi-infinite bodies is [2]

$$p(h) = \frac{8\Delta\gamma}{3\varepsilon}\left[\left(\frac{\varepsilon}{h}\right)^3 - \left(\frac{\varepsilon}{h}\right)^9\right], \tag{1}$$

where h is the gap between two surfaces, ε is the equilibrium distance between them and, $\Delta\gamma$ is the surface energy.

If the curvature of the surface is very small and the distance between the two surfaces is small, Eq. (1) is used to obtain the local pressure (Derjaguin's approximation [15]).

Using the parabolic approximation $h = \alpha + r^2/2R$ for the gap, where R is the equivalent radius of curvature, $1/R = 1/R_1 + 1/R_2$, and α is the approach distance, the total force between two bodies is simply [16]

$$w = \frac{8\pi R\Delta\gamma}{3}\left[\left(\frac{\varepsilon}{\alpha}\right)^2 - \frac{1}{4}\left(\frac{\varepsilon}{\alpha}\right)^8\right]. \tag{2}$$

Jump-to-contact occurs when the gradient of the force-approach relation equals the stiffness of the cantilever, k:

$$k = \frac{dw}{d\alpha}. \tag{3}$$

When only the van der Waals force is considered, this gives the Israelachvili and Tabor [3] equation

$$\left(\frac{\varepsilon}{\alpha}\right)^3 = \frac{3k\varepsilon}{16\pi R\Delta\gamma} \tag{4}$$

(where the Hamaker constant, H_A, used in [3] has been replaced by $H_A \equiv (16\pi/3)\ \varepsilon^2\Delta\gamma$). When the full Lennard-Jones interaction is used, the corresponding equation is

$$\left(\frac{\varepsilon}{\alpha}\right)^3 - \left(\frac{\varepsilon}{\alpha}\right)^9 = \frac{3k\varepsilon}{16\pi R\Delta\gamma}. \tag{5}$$

2.2. The Adhesive Elastic Problem

The adhesive elastic contact problem has been described in detail by Greenwood [2], and effective methods of performing the numerical solution described also by Greenwood [17]: here only a brief summary is given. Using the parabolic approximation $h = \alpha + r^2/2R$ for the initial gap and the half-space approximation for the deformation, the gap between the surfaces of two elastic spheres is [2]

$$h(r) - \alpha - \frac{r^2}{2R} - \frac{4}{\pi E^*}\int_0^\infty \frac{p(s)s}{r+s} K\left(\frac{2\sqrt{rs}}{r+s}\right) ds = 0, \tag{6}$$

where E^* is the contact modulus, $\frac{1}{E^*} = \frac{1-\nu_1^2}{E_1} + \frac{1-\nu_2^2}{E_2}$, r, s are the radial coordinates, and $K(\cdot)$ is the complete elliptic integral of the first kind.

The pressure $p(s)$ is obtained from Eq. (1), but only after the gap thickness, $h(s)$, has been found from Eq. (3), so that an iterative method is required. Note the convention that the two spheres are "in contact" when the pressure vanishes, so at this point the approach α is ε, not zero. Integrating the pressure gives the total force, w, as

$$w = 2\pi \int prdr. \tag{7}$$

Equations (2) to (7) apply equally to the crossed mica cylinders geometry of Israelachvili and Tabor, and to adhesive contact between an AFM tip and a surface.

When the following non-dimensional variables are used,

$$H = \frac{h}{\varepsilon} \quad A = \frac{\alpha}{\varepsilon} \quad P = \frac{p\varepsilon}{\Delta\gamma} \quad W = \frac{w}{2\pi R\Delta\gamma},$$

the solution of Eq. (6) depends only on the Tabor parameter[1] [18,19] $\mu = \left(\frac{R\Delta\gamma^2}{E^{*2}\varepsilon^3}\right)^{1/3}$.

$$H(\hat{r}) - A - \frac{1}{2}\hat{r}^2 + \frac{32\mu^{3/2}}{3\pi}\int_0^\infty \left[\frac{1}{H(\hat{s})^3} - \frac{1}{H(\hat{s})^9}\right]\frac{\hat{s}}{\hat{r}+\hat{s}}K\left(\frac{2\sqrt{\hat{r}\hat{s}}}{\hat{r}+\hat{s}}\right)d\hat{s} = 0. \quad (8)$$

In 1992, Attard and Parker [10] assumed that the deformation varies slowly compared with the curvature:

$$u(r) \approx u(0).$$

Based on this assumption, they gave an analytical formula for jump criterion for the fixed-grips device. Their formula is based on the deformed surface. The criterion can be expressed as $H(0) = 1.84\,\mu^{7/3}$.

3. JUMP-TO-CONTACT DISTANCE

3.1. Rigid Solids and Low Tabor Parameter Surfaces

Figure 2 shows the force-approach curves for the adhesive contact between two spheres. The solid line is for the *van der Waals* force between *rigid* spheres: the dotted line is for the *Lennard-Jones* force between *rigid* spheres. The other lines are for the *Lennard-Jones* force between *deformable* spheres with Tabor parameters $\mu = 0.01$, 0.1, 0.2, and 0.5. For all these cases, the force-approach curve has not developed the S-shape shown in Fig. 1, so there is no jump-in-contact for a fixed grips device in these cases. All the approaching parts of the force-approach curves are very similar to the van der Waals curve for $\mu \leq 0.1$. The simple relation [Eq. (4)] for the *van der Waals* force between *rigid* spheres can predict the jump-to-contact distance well for $\mu \leq 0.1$ when the stiffness of the cantilever is less than the maximum gradient of the force-approach curve.

[1]Tabor [18] proposed this parameter which can explain the discrepancies between the JKR [9] and the Bradley [16] models. Muller *et al.* [19] subsequently performed a numerical simulation and confirmed that Tabor's parameter does govern the transition.

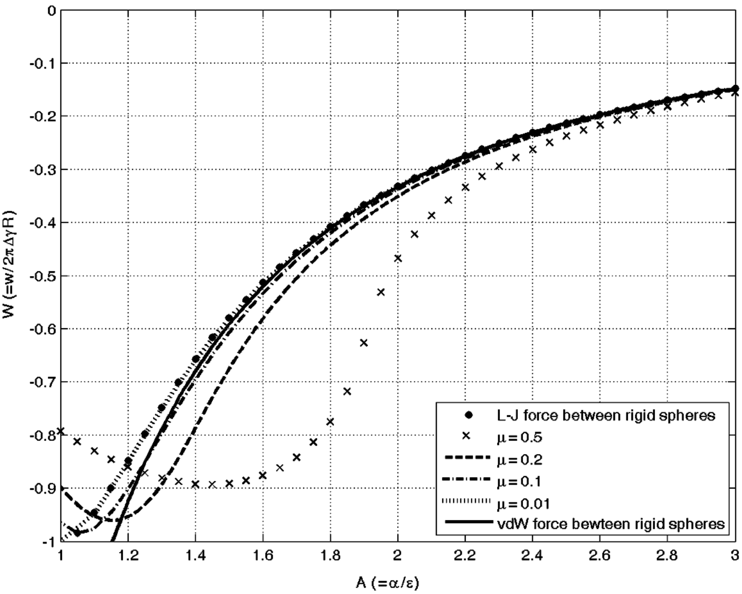

FIGURE 2 The force-approach relation for a small Tabor parameter.

3.2. Jump-to-Contact for Large Tabor Parameter

3.2.1. The van der Waals Force Between Deformable Spheres

For large Tabor parameter ($\mu = 3$, 5, and 10), the approaching part of the force-approach curve is shown in Fig. 3. These curves have similar shapes. In this figure, $A > 6$ for $\mu = 3$, $A > 9$ for $\mu = 5$, and $A > 16$ for $\mu = 10$. Therefore, $H^9 >> H^3$ (where A, $H > 2.5$) for these cases and the *van der Waals* force dominates the interactions. If the van der Waals force is the only force, the gap between the surfaces of two elastic spheres is

$$H(\hat{r}) - A - \frac{1}{2}\hat{r}^2 + \frac{32\mu^{3/2}}{3\pi}\int_0^\infty \frac{1}{H(\hat{s})^3}\frac{\hat{s}}{\hat{r}+\hat{s}}K\left(\frac{2\sqrt{\hat{r}\hat{s}}}{\hat{r}+\hat{s}}\right)d\hat{s} = 0. \qquad (9)$$

In Eq. (9), the Tabor parameter can be omitted by the following non-dimensional parameters.[2]

$$H' = \frac{H}{\mu^{3/7}} \quad A' = \frac{A}{\mu^{3/7}} \quad K' = \frac{K}{\mu^{3/7}}$$
$$\hat{r}' = \frac{\hat{r}}{\mu^{3/14}} \quad \hat{s}' = \frac{\hat{s}}{\mu^{3/14}}$$

[2]H and A have the same "unit." $\hat{r}$ and $\hat{s}$'s unit is $H^{1/2}$. The unit for Eq. (9) is $O(H) + O(\frac{\mu^{3/2}H^{1/2}}{H^3}) = 0$. Thus, by setting $H' = \frac{H}{\mu^{3/7}}$, the Tabor parameter, μ, can be omitted.

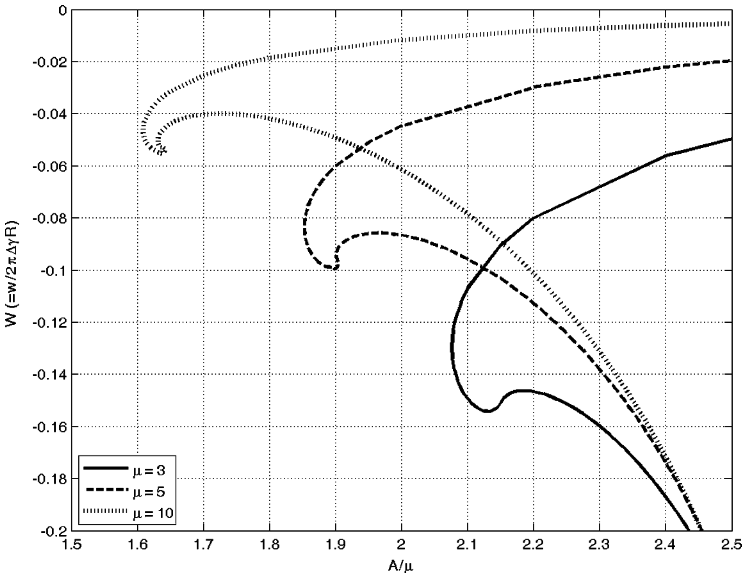

FIGURE 3 The approach part of the force-approach curve with Tabor parameter $\mu = 3$, 5, and 10.

Thus, Eq. (9) becomes

$$H'(\hat{r}') - A' - \frac{1}{2}\hat{r}'^2 + \frac{32}{3\pi}\int_0^\infty \frac{1}{H'^3}\frac{\hat{s}'}{\hat{r}' + \hat{s}'}K\left(\frac{2\sqrt{\hat{r}'\hat{s}'}}{\hat{r}' + \hat{s}'}\right)d\hat{s}' = 0. \tag{10}$$

The solution is independent of the Tabor parameter.

Figure 4 shows the relationship between A' and W'. It is found that all the curves collapse into one curve. That is, for $A > 2.5$, the solution with the van der Waals force is a good approximation for the solution with the Lennard-Jones potential.

The new non-dimensional total force can be defined as

$$W' = \frac{8}{3}\int \frac{\hat{r}' d\hat{r}'}{H'^3} = W\mu^{6/7}. \tag{11}$$

The new non-dimensional spring constant is defined as

$$K' = \frac{dW'}{dA'} = \frac{\mu^{9/7} dW}{dA} = \mu^{9/7}K. \tag{12}$$

3.2.2. Empirical Formulas

The numerical simulation is cumbersome. If one wishes to compare the result with experiment data, a simpler equation that approximates

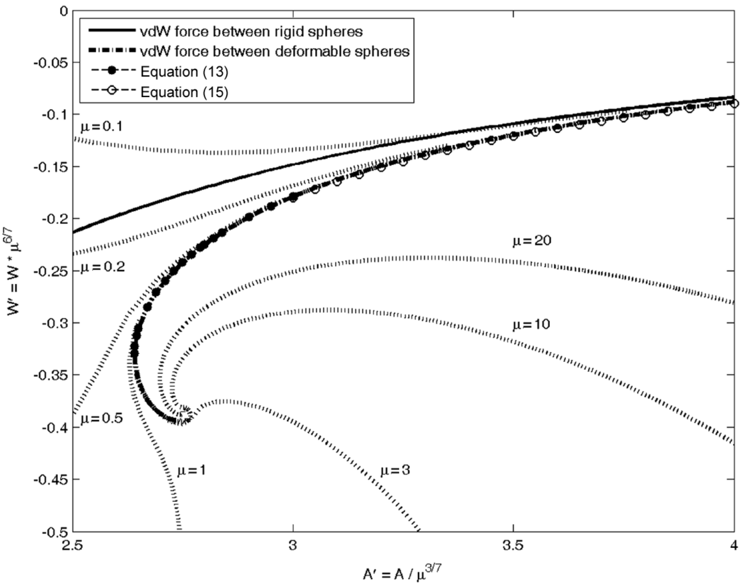

FIGURE 4 The approach part of the force-approach curves with new non-dimensional parameters. The lines for Eqs. (13) and (15) nearly coincide with that for the *van der Waals* force between *deformable* spheres.

the jump-to-contact distance and the total force is preferred. It is not practical to make a formula fitting the curve for the jump-to-contact distance and the total force for all different Tabor parameters. Thus, the equation for the van der Waals force is proposed.

The force-approach curve up to the vertical tangent is related to jump-to-contact. Using curve-fitting, the exact relation between A' and W' can be approximated as below:

1. For $A' \leq 3$, where $K' \geq 0.1766$,

$$A' = 206.2W'^4 + 249.9W'^3 + 126.8W'^2 + 31.72W' + 5.835 \qquad (13)$$

$$K' = \frac{1}{824.8W'^3 + 749.7W'^2 + 253.6W' + 31.72} \qquad (14)$$

2. For $4 \geq A' > 3$, where $0.0487 \leq K' < 0.1766$,

$$W' = -\exp\{0.0806A'^4 - 1.234A'^3 + 7.209A'^2 - 19.61A' + 19.026\} \qquad (15)$$

$$K' = W'\{0.3224A'^3 - 3.702A'^2 + 14.418A' - 19.61\} \tag{16}$$

3. For $5 \geq A' > 4$, where $0.0213 \leq K' < 0.0487$,

$$W' = -\exp\{0.0047A'^4 - 0.1006A'^3 + 0.8475A'^2 - 3.718A' + 4.1065\} \tag{17}$$

$$K' = W'\{0.0188A'^3 - 0.3018A'^2 + 1.6950A' - 3.718\} \tag{18}$$

4. For $A' > 5$, where $K' < 0.0213$,

$$W' = -\frac{4}{3}A'^{-2} \tag{19}$$

$$K' = \frac{8}{3}A'^{-3} \tag{20}$$

Equations (13) to (18) can approximate the case for the *van der Waals* force between *deformable* spheres with the errors of A' and K' less than 1%. Equations (19) and (20) can approximate the case for the *van der Waals* force between *deformable* spheres with the errors of A' and K' less than 2%.

Figure 5 shows the curves of non-dimensional spring constant, K', *versus* non-dimensional jump-to-contact distance, A'. For $\mu \geq 1$, all curves nearly coincide. For $\mu = 0.5$, the *van der Waals* force between *deformable* spheres can obtain the jump-to-contact distance for $A' \geq 3$ ($A \geq 2.22$). For $\mu = 0.2$, the *van der Waals* force between *deformable* spheres can obtain the jump-to-contact distance for $A' \geq 3.8$ ($A \geq 1.91$). For $\mu = 0.1$, the jump-to-contact can be obtained by the *van der Waals* force between *rigid* spheres for $A' \geq 3.8$ ($A \geq 1.42$).

If the spring constant is given, the jump-to-contact distance can be obtained. The procedure for finding the jump-to-contact distance is:

1. Find the Tabor parameter, μ.
2. Find the non-dimensional spring constant, K and K', by Eqs. (12) and (19).
3. If $\mu \leq 0.1$ or $K' < 0.0213$, the jump-to-contact distance can be obtained by Eq. (15). Otherwise, go to Step 4.
4. From the spring constant, K', find the adequate interval and find the jump-to-contact distance, A'.

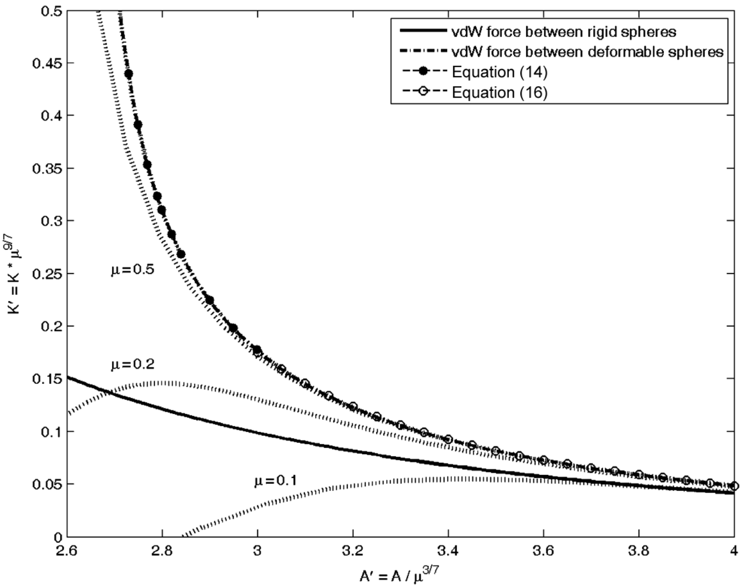

FIGURE 5 Non-dimensional spring constants *versus* jump-to-contact distance. The line for the *van der Waals* force between *deformable* spheres nearly coincides with those for Eqs. (14) and (16). The lines for the *Lennard Jones* force between *deformable* spheres with Tabor parameter $\mu = 1$, 3, 10, and 20 (not shown in this figure) coincide with that for the *van der Waals* force between *deformable* spheres.

3.3. Jump-to-Contact for Fixed Grips

In the approaching part, all $A' - W'$ curves for large Tabor parameters collapse into the curve for the van der Waals force. The jump-to-contact distance for the van der Waals force can be used to obtain the one for large Tabor parameters.

In the case of *van der Waals* force between *deformable* spheres, for a fixed grips apparatus ($K' = \infty$), the jump-to-contact distance occurs at the vertical tangents in Fig. 4:

$$A' = 2.641. \tag{21}$$

That is, the jump-to-contact distance for a fixed grips apparatus and large Tabor parameter is:

$$A = 2.641\mu^{3/7}. \tag{22}$$

Figure 6 shows the jump-to-contact distances from the numerical simulation with the Lennard-Jones solution and with the van der

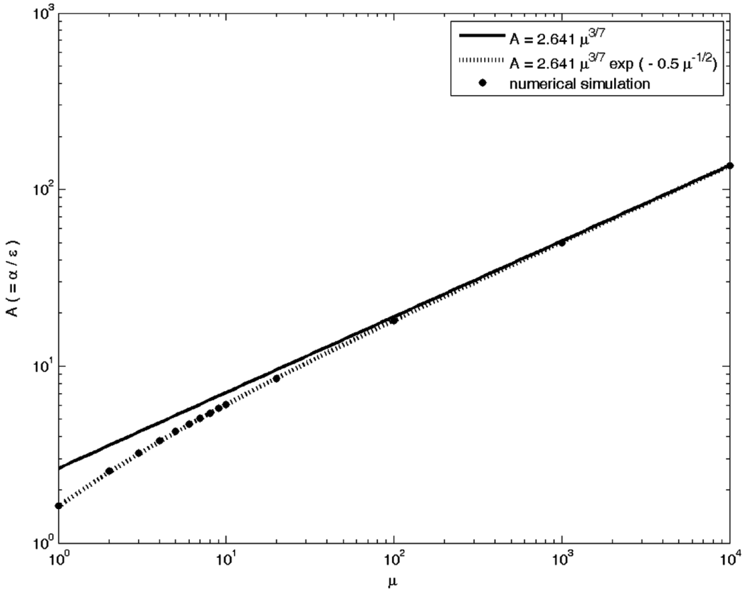

FIGURE 6 Jump-to-contact distances with the Lennard-Jones potential and those obtained by Eqs. (22) and (23).

Waals force. A good curve fit for jump-to-contact distance with the Lennard-Jones potential and a fixed-grips device is:

$$A = 2.641\mu^{3/7}\exp\left(-\frac{1}{2\sqrt{\mu}}\right). \tag{23}$$

4. DISCUSSION

The formulae proposed in Section 3 can obtain the jump-to-contact distance for large Tabor parameters, such as an ATM tip measuring a membrane or cells. Butt *et al.* [20] argued that the jump-to-contact can be avoided by using a stiff cantilever. From Section 3, it is obvious that the jump-to-contact phenomena can not be avoided for large Tabor parameters. The only method for avoiding jump-to-contact for large Tabor parameters is to keep the distance larger than the jump-to-contact distance.

Although other forces are not considered in this paper, the method in Section 3 can still be applied, as long as the van der Waals force dominates the interaction between the tip and the surface.

Israelachvili and Tabor's formula can predict the jump-to-contact distance for $\mu \leq 0.1$ or $K' < 0.0213$. The result in Section 3 can obtain the jump-to-contact distance for $A \leq 2.5$ and for all other cases.

Attard and Parker's formula is based on the deformed surface [$h(0)$] and is for the fixed grips device only. For predicting the jump-to-contact distance, using α is better than $h(0)$, since the instrument can detect the position of α, but not of $h(0)$. Therefore, the formulae in Section 3 are better than Attard and Parker's formula in predicting the jump-to-contact phenomena.

5. EXPERIMENTAL

In 2006, Wahl *et al.* [21] used the AFM to measure the force between a polydimethylsiloxane (PDMS) surface and a tip. In Fig. 7, the tip jumps to contact at point A, whose position is about 585 nm. The tip jumps off contact at point B, whose position is about 655 nm. The distance between jump-to-contact and jump-off-contact is about 70 nm.

In this case, $\Delta\gamma = 49\,\mathrm{mJ/m^2}$, $R = 10\,\mu\mathrm{m}$, $\varepsilon = 0.5\,\mathrm{nm}$. Wahl *et al.* [21] show that the Tabor parameter ($\mu = 96.28$) for their contacts is high

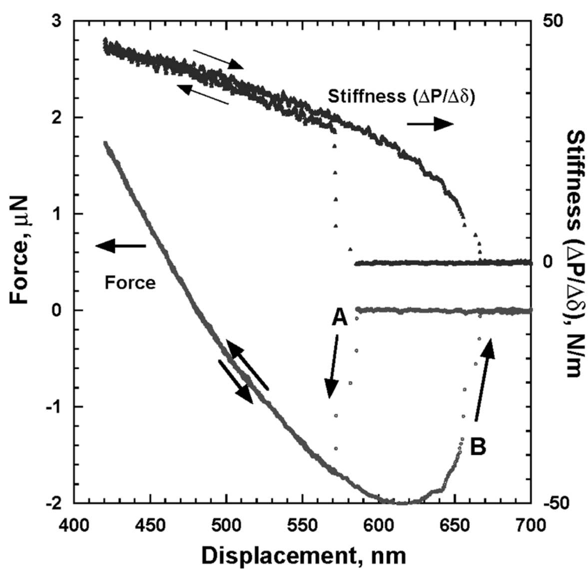

FIGURE 7 Distance *vs* force (lower) and stiffness (upper) curves for PDMS obtained with a 10-μm radius tip at an approach/retract rate of 1 nm/s (Reprinted from [21] with permission from Elsevier).

enough to expect JKR behaviour. For a fixed-grips device with a large Tabor parameter, the jump-off-contact distance can be predicted by the JKR model [9,14].

$$\frac{A}{\mu} = \left(\frac{9\pi}{8}\right)^{2/3} 3^{-1/3}$$

$$A = 154.89$$

$$\alpha = 77.45\,\mathrm{nm}$$

Since the experimental set up approaches the condition of "fixed grips" or an infinite stiffness apparatus [21], the jump-to-contact distance can be obtained by Eq. (22).

$$A = 2.641\mu^{3/7} = 18.70$$

$$\alpha = 9.35\,\mathrm{nm}$$

The distance between jump-to-contact (9.35 nm) and jump-off-contact (77.45 nm) is 68.1 nm, which is similar to that of the experiment (70 nm).

Israelachvili and Tabor [3] did not predict a jump into contact ($\alpha = 0$). This conflicts with observation as in Fig. 7.

6. CONCLUSION

The force-approach relation for the adhesive contact based on the Lennard-Jones potential with the Derjaguin approximation is investigated. In the approaching part, the solution with the *van der Waals* force between *deformable* spheres can approximate that with the Lennard-Jones potential for $A \geq 2.5$. Empirical formulae for the approaching part of the force-approach curve are proposed. By using the empirical formulae, the jump-to-contact distance can be obtained. For a fixed grips device, the jump-to-contact distance is also obtained for large Tabor parameters. The formulae in Section 3 fit well with one set of published data, so they may be useful if more experimental data are available for further comparisons in the future.

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