

RESEARCH ARTICLE | AUGUST 16 2021

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Appl. Phys. Lett. 119, 071603 (2021)

<https://doi.org/10.1063/5.0059176>

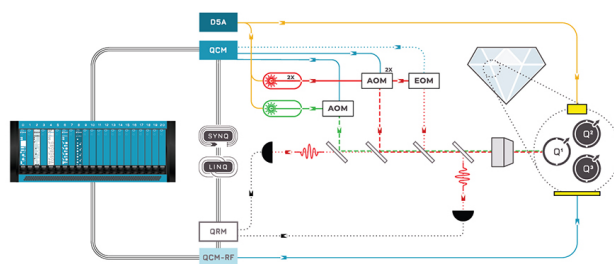


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Cite as: Appl. Phys. Lett. **119**, 071603 (2021); doi: [10.1063/5.0059176](https://doi.org/10.1063/5.0059176)

Submitted: 7 June 2021 · Accepted: 3 August 2021 ·

Published Online: 16 August 2021



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ABSTRACT

Understanding the adhesion between rough surfaces has practical significance. We derive a simple analytical formula on the basis of the classic order statistics to predict the interfacial binding energy between rough surfaces. It is found that the strong length scale dependence of adhesion ranging from the nominal size scale down to any artificially defined cutoff length scale in constructing a rough profile can be considered as a purely statistic performance resulted from different samplings and can be further described by a single parameter called sampling number. We compare the formula predictions with the experimental results and demonstrate that our simple formula holds its accuracy especially for the Derjaguin–Muller–Toporov adhesion case.

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Adhesion between rough solids shows strong length scale dependence (e.g. microscopic objects are always much stickier than those in a macroscopic scale)¹ and is highly sensitive to the surface topography (roughness).^{2–4} Understanding the effect of surface roughness on adhesion has important practical implications especially in the fields of thin film coating,^{5,6} powder technology,⁷ micro-electromechanical systems (MEMS),^{8,9} and engineered mimics.^{10–12} Two types of theories, taking the name of Hamaker,¹³ and Johnson–Kendall–Roberts (JKR)¹⁴ or Derjaguin–Muller–Toporov (DMT),¹⁵ are the basis for the development of the adhesion model. The Hamaker theory uses a simple pairwise integration approach under the continuum approximation to estimate the interfacial binding energy (or work of adhesion), while the JKR or DMT theory considers the influence of both material property and surface energy on the adhesion behavior. Note that, both theories are initially derived under smooth setting and, therefore, may overestimate the adhesion measured in experiments where surface roughness always exists.

Various of theoretical studies by numerical or analytical approaches have been proposed for the past several decades, resulting in an intensive understanding of the effect of surface roughness on adhesion. However, numerical approaches, such as molecular dynamics,^{16,17} boundary element,^{18,19} finite element,^{20,21} Monte Carlo

method,²² and multi-level integration technique,²³ are not computationally economical, because of the requirement of multiple calculations to yield appropriate averages. Persson has derived an analytical theory on the basis of the finding that the interfacial stress distribution satisfies the diffusion-type equation.²⁴ Although Persson's theory is powerful for the investigation of an adhesive (or non-adhesive) contact between rough solids at different magnifications, it may be difficult to lead to a simple form of solution for a general case due to a set of integration equations in it.²⁵

In this Letter, we use the Hamaker's additive approach under the framework of classic order statistics to derive a simple analytical formula for predicting adhesion between rough surfaces. Hamaker's approach that ignores the deformation effect may be questionable in contact mechanics but is still sufficiently accurate especially in estimating interfacial binding energy between elastically hard materials.^{26,27} Our model gives a clear relation between surface adhesion and roughness at different length scales and shows good agreement with the experiments. More importantly, it is revealed that the length scale dependence of adhesion also has statistic characteristic corresponding to a single parameter, sampling number, with independence of the surface roughness.

Our model starts from recent findings reported by Pastewka and Robbins that the contact of the first asperity leads to large variation in adhesion prediction between different realizations of random surfaces.¹⁷ This result can be well interpreted by the classic order statistics as follows: The profile of a rough surface may have a discrete form of height function (one sampling) with a given distribution, constant probability parameter (e.g., standard derivation σ), and a finite sampling number (M); if the first contact forms at the highest profile, and supposing that this contact may be important, the entirety adhesion should be also a random variable with a distribution governed by both the height function and the statistical feature of the maximum value in each sampling. Note that, the height function for all samplings can follow the same distribution, but the maximum height for each one may be different.

The interaction between two rough surfaces can be mapped to that between flat and rough surfaces, since the relative (local gap) rather than the absolute profile governs a contact problem.² Figure 1 illustrates the geometry of our model, in which there is an infinite flat solid on the top and a rough finite one on the bottom. The profile height (function) of the rough surface is $z(x)$, where x denotes the lateral coordinate with a tunable form of vector corresponding to the dimension of the problem. The coordinate is located at zero means ($\bar{z} = 0$) with positive value in the upwards direction. Under the continuum approximation, the rough solid can be considered as a combination of a set of volume elements closely packed in a total sampling number of M .²⁸ The value of M directly reflects the length scale of the rough surface in two limit ways: for a given size of the volume element, M determines the maximum size, i.e., the nominal size (area for a 2D surface, while width for 1D), while when the nominal size is fixed, M measures the discrete degree, i.e., the resolution in the lateral direction, τ . If the potential energy between a volume element and an infinite flat solid with a distance d is $U_0(d)$, the potential energy of the assembly is, thus, given by the convolution,^{4,29} as

$$U(g) = \int_{\mathbb{R}} U_0(g - z)f(z)dz, \quad (1)$$

where \mathbb{R} is the set of the real number, g is the distance between the reference plane and the upper flat surface, and $f(z)$ is the probability density function of the profile height (PDF). Since truncation is not considered in Hamaker's approach, U reaches its maximum value as the highest profile z_{\max} contacts with the upper surface ($g = z_{\max}$; the

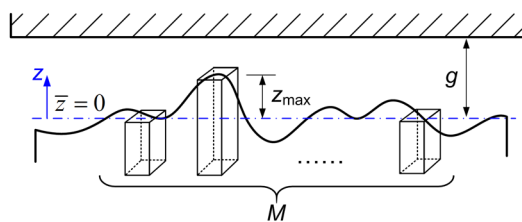


FIG. 1. Schematic illustration of geometry in our model. The top solid is ideally flat and infinitely extended, and the bottom one has a rough surface with a profile height function $z(x)$. z_{\max} denotes the highest profile. The gap between two solids g is defined as the distance from the upper surface to a reference plane of zero means, i.e., $\bar{z} = 0$. Under the continuum approximation, the lower solid is divided into a set of volume elements in a total number of M , called sampling number.

potential energy is generally negative, but in the present context, it is positive for convenience). Thus, the effective interfacial binding energy with the presence of surface roughness can be expressed according to its definition as: $\gamma_{\text{eff}} = U(z_{\max})$.³ As z_{\max} is also a random variable that may be changed for different samplings, we can use the expectation value of the γ_{eff} to measure its mean level, i.e.,

$$\mathbb{E}(\gamma_{\text{eff}}) = \mathbb{E}[U(z_{\max})] = \int_{\mathbb{R}} (U|_{z_{\max} = x})f_{z_{\max}}(x)dx, \quad (2)$$

where $\mathbb{E}(\dots)$ denotes the expectation and $f_{z_{\max}}(x)$ is the PDF of the z_{\max} , which is given according to the order statistics by³⁰

$$f_{z_{\max}}(x) = Mf(x) \left[\int_{-\infty}^x f(\tau)d\tau \right]^{M-1}. \quad (3)$$

Combining Eqs. (1)–(3), we have

$$\mathbb{E}(\gamma_{\text{eff}}) = M \int_{\mathbb{R}} \left[\int_{\mathbb{R}} U_0(x - \eta)f(\eta)d\eta \right] \left[\int_{-\infty}^x f(\tau)d\tau \right]^{M-1} f(x)dx. \quad (4)$$

Equation (4) shows a general form of $\mathbb{E}(\gamma_{\text{eff}})$, which can be determined by three factors: potential energy between two flat solids $U_0(d)$, PDF of the profile height $f(z)$, and the sampling number M . The first two factors, which are often considered in previous investigations,³ mathematically describe how the two solids interplay with each other with the presence of the surface roughness, while the last one is corresponding to the length scale. (We will discuss it later in detail.) In principle, both U_0 and $f(z)$ can have an arbitrary form, e.g., with U_0 being derived from the van der Waals model for pure adhesion,²² the Dugdale model for a constant attractive traction,³¹ or the Lennard-Jones (LJ) model for a more canonical case,³² and with $f(z)$ being either Gaussian or non-Gaussian.^{22,33} Here, we only show representative results for the U_0 of LJ potential together with a Gaussian distribution of $f(z)$. If the energy potential between two atoms follows the 6–12 LJ law, $U_0(d)$ can be obtained by the integral over the half space, thus given by³¹

$$U_0(d) = \frac{4\gamma}{3} \left[\left(1 + \frac{d}{d_c}\right)^{-2} - \frac{1}{4} \left(1 + \frac{d}{d_c}\right)^{-8} \right], \quad (5)$$

where γ is the interfacial binding energy in an ideal condition (no surface roughness) and d_c denotes the characteristic distance of the interaction. For the Gaussian distribution with zero means, we have the PDF of

$$f(\tau) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{\tau^2}{2\sigma^2}\right). \quad (6)$$

Equations (4)–(6) are computationally efficient for extensive parameter studies. Figure 2 presents the normalized expectation of the effective interfacial binding energy, $\bar{\mathbb{E}}(\gamma_{\text{eff}}) = \mathbb{E}(\gamma_{\text{eff}})/\gamma$, as functions of the normalized surface roughness, $\bar{\sigma} = \sigma/d_c$, for different values of M . Both of $\bar{\sigma}$ and M are taken over several orders of magnitude for the purpose of showing a general dependence of interfacial adhesion on them with practical significance for various engineering applications.^{34,35} Specifically, the maximum value of M is set up to 10^5 to cover most characterizations of surface roughness using precise

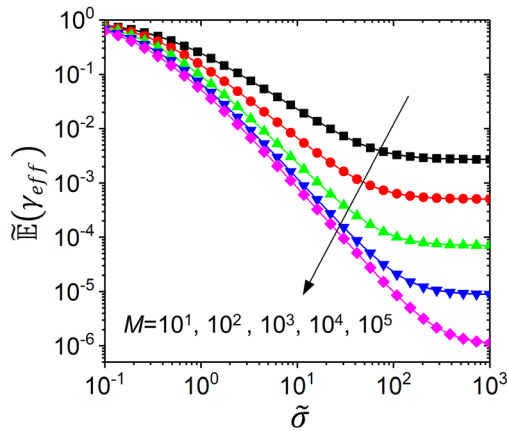


FIG. 2. Plot of normalized expectation of effective interfacial binding energy $\tilde{\mathbb{E}}(\gamma_{eff}) = \mathbb{E}(\gamma_{eff})/\gamma$ as functions of normalized surface roughness $\tilde{\sigma} = \sigma/d_c$ for different values of sampling numbers, M . The downwards arrow indicates the increase trend of M from 10^1 to 10^5 . Adhesion strongly depends not only on the surface roughness but also on the specific sampling method.

instrument (e.g., atomic force microscopy for a nano- to micro-scale sample,^{9,36} and a stylus profilometer for a micro- to millimeter scale³⁷) with a limited resolution usually no more than 1000 pixels per scan line (i.e., total data points $\sim 10^5$ – 10^6). $\tilde{\mathbb{E}}(\gamma_{eff})$ dramatically decreases from ~ 1 to $\sim 10^{-2}$ – 10^{-3} as $\tilde{\sigma}$ increases from 10^{-1} to 10^1 , implying the huge influence of the surface roughness on the adhesion. As $\tilde{\sigma}$ continues to increase up to 10^2 – 10^3 , the decrease trend almost vanishes and $\tilde{\mathbb{E}}(\gamma_{eff})$ reaches a plateau value of 10^{-3} for $M = 10$ and of 10^{-6} for $M = 10^5$. This is because, when $\tilde{\sigma}$ reaches up to 10^2 – 10^3 (a surface roughness is of 2–3 order of magnitude larger than the characteristic distance of the interaction), the number of volume element having comparable potential energy with γ could be very small. In such a condition, the adhesion behavior loses its statistic characteristic, thus showing independence of the surface roughness.

It is also found in Fig. 2 that in the whole range of $\tilde{\sigma}$, larger M leads to smaller $\tilde{\mathbb{E}}(\gamma_{eff})$. This is true, because more sampling numbers lead a rough surface to have a higher probability of a larger z_{max} , thus to show a weaker adhesion. For a better understanding of the role of their correlation on the length scale issue, in Fig. 3, we first assume that there is an infinitely extended rough surface with a continuous and random profile height z , which is further assumed to follow a given PDF (surface i, $M_i \rightarrow \infty$). Any random realization for generating a rough surface under the same PDF can be considered as one sampling on surface i with finite sampling number (surface ii, $M_{ii} \ll M_i$). As one of the two ways that can result in the increase in M is to increase the nominal size of the surface at a fixed resolution (surface iii, larger than surface ii, and $M_{iii} > M_{ii}$), the rough surface of larger nominal size is expected to show a weaker adhesion in comparison with that of smaller one. This prediction is the well-known length scale dependence that microscopic objects are often sticky, while those in macroscopic are not.^{1,2} Many current theories explain this phenomenon by showing that larger objects are rougher than smaller ones.^{1,4,25} According to our model, however, it is also a statistic result of different samplings with independence of the surface roughness, since the PDF for the generation of surfaces ii and iii are unchanged.

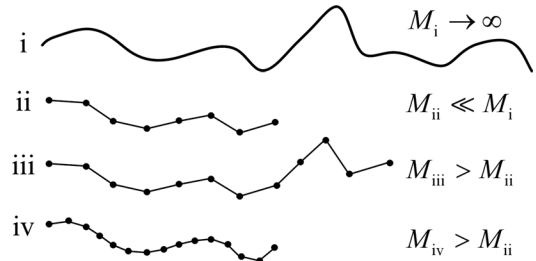


FIG. 3. Schematic illustration of different samplings on an infinitely extended and randomly rough surface (surface i, $M_i \rightarrow \infty$) with a continuous profile height function z and a probability density function $f(z)$. Any random realization for the generation of a rough surface with the same $f(z)$ can be considered as a sampling with finite M on surface i. Surface ii is an example of a sampling ($M_{ii} \ll M_i$). Surface iii has larger nominal size, while surface iv has higher resolution in comparison with surface ii. Both two ways can lead to the increase in M ($M_{iii} > M_{ii}$, and $M_{iv} > M_{ii}$). Since surface i is supposed to be infinitely large, while surfaces ii, iii, and iv are finite, the former one is drawn with longer contour length than the others.

In other words, even if the surface roughness is a constant, the adhesion could decrease with the increase in the nominal size.

In addition to the nominal size, the resolution can also determine the value of M (surface iv, with the same length of surface ii but higher discrete degree, also $M_{iv} > M_{ii}$), which means that the adhesion could depend on the discrete degree of the rough surface in modeling. This seems not correct, because any physical quantity should not depend on an artificially defined parameter, i.e., adhesion behavior of surface ii should be nearly as the same as that of surface iv. In fact, the resolution determines the cutoff length below which the fine scale surface roughness is neglected. For modeling adhesion at a lower resolution, as reported by Joe *et al.*, the potential energy $U_0(d)$ in Eq. (5) should be modified to a weaker one, so as to allow for the effect of the neglected fine scale roughness.²⁹ If one model uses an identical form of $U_0(d)$, it can overestimate the adhesion in a lower resolution or underestimate that in a higher one [e.g., lower $\tilde{\mathbb{E}}(\gamma_{eff})$ for larger M in this study]. Our formula reveals, from the point of view of statistics, the importance of a proper effective potential function in achieving an accurate result at a specific resolution. In short, adhesion between rough surfaces shows strong nominal size dependence and needs to carefully deal with at different resolutions in modeling. These two phenomena can be unified to a simple relationship between $\tilde{\mathbb{E}}(\gamma_{eff})$ and the sampling number, M [see Eq. (4)] and, thus, can be both defined as a length scale effect behaving in two limit scales.

Finally, in order to assess the accuracy of our formula, Fig. 4 compared our results with the experiments in several conditions.^{38–41} Generally, adhesion experiments are carried out in a sphere-to-flat configuration. The effective interfacial binding energy γ_{eff} can then be evaluated from the measured pull-off force F and the radius of the sphere R , by $\gamma_{eff} = F/\alpha\pi R$, where α is a modal variable with $\alpha = 2$ for the DMT case of $\mu \ll 1$, and $\alpha = 1.5$ for the JKR case of $\mu \gg 1$, $\mu = (\gamma^2 R/E^* d^3)^{1/3}$ is the Tabor constant determining which type of the contact model is more accurate in describing adhesion behavior, and E^* is the composite Young’s modulus.⁴² As both the surfaces in experiments are rough, the composite roughness, $\sigma^* = (\sigma_s^2 + \sigma_f^2)^{1/2}$, is used as the horizontal axis, where the subscripts “s” and “f” denote

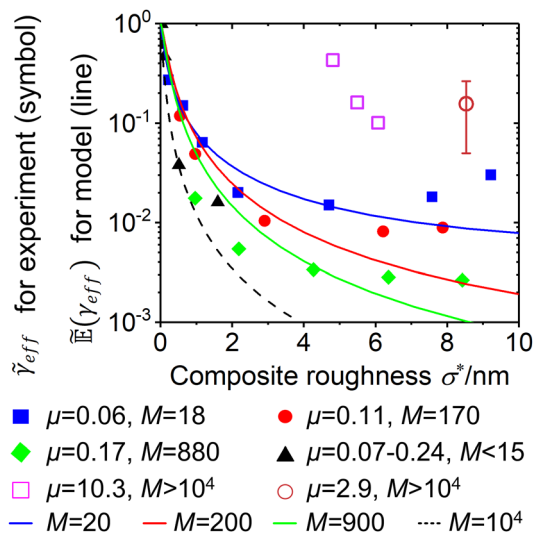


FIG. 4. Comparison between the present model and the experimental data reported in the literature by plotting adhesion as a function of the surface roughness. The vertical axis denotes $\tilde{E}(\gamma_{eff})$ for our model results (lines), while denotes $\tilde{\gamma}_{eff}$ for the experimental data (symbols). The horizontal axis is the composite roughness of the two surfaces in experiments. The solid symbols stand for the DMT case, while the open ones for the JKR case. The Tabor constant μ and sampling number M for these data are listed at the bottom of the figure.

the sphere and nominal flat surface, respectively. For the experimental data (symbols), the vertical axis denotes $\tilde{\gamma}_{eff} = \gamma_{eff}/\gamma$, which can be further calculated approximately by $\tilde{\gamma}_{eff} \approx F/\alpha\pi R\gamma_{\sigma^* \approx 0}$, where $\gamma_{\sigma^* \approx 0}$ is the γ measured at $\sigma^* \approx 0$. ($\sigma^* = 0$ is experimentally impracticable, thus the data measured at the minimum roughness are used as the normalization quantity.) For the results predicted by our model (lines), the vertical axis denotes $\tilde{E}(\gamma_{eff})$. A proper value of M can be estimated by comparing the nominal area and the resolution of the rough surface in contact with $M \approx \pi(a/\tau)^2$, where a is the contact radius for adhesion instability that can be calculated according to the DMT or JKR theory as $a_{DMT} = (3\pi\gamma_{eff}R^2/2E^*)^{1/3}$ or $a_{JKR} = 3^{1/3}a_{DMT}$, and τ is the resolution. In a more general consideration, M also depends on the high moment information of the rough surfaces, which may give different distributions but even share the same mean or variance. Packaging this information in M could, however, largely increase the complexity of our main formula [Eq. (4)] and, thus, is not concerned in this study. In Fig. 4, symbols with $\mu < 1$ are solid (the DMT case), while those with $\mu > 1$ are open (the JKR case). (Data are presented in Table S1 in detail, the [supplementary material](#).) The solid lines with $M = 20, 200,$ and 900 are very close to the solid symbols with $M = 18, 170,$ and 880 , respectively, implying the sufficient accuracy of our model for the DMT case. Since materials in many applications involving bonding or debonding control on microscopic objects are elastically hard (e.g., silicon in MEMS,^{8,9} polystyrene or silica particle in self-assembly,⁴³ and diamond in a scanning probe technique^{39,44}) and may belong to the DMT case, our model with a simple formula holds its power in deriving a general adhesion rule for guiding the practice. On the other hand, the great underestimation of the dotted line with

$M = 10^4$ in comparison with the open symbols with $M > 10^4$ means the feasibility loss of our model for the JKR case. This is because the deformation is assumed as not important in our model for the purpose of deriving a simple formula. Such assumption may have neglectable influence for the DMT case, where the surface energy induced tractions are also assumed to not alter the contour of the contact body.²⁶ However, for the JKR case, deformation is important, and thus, our model cannot be applied to this case.

In summary, we derived a simple formula on the basis of the classic order statistics for the estimation of interfacial binding energy with the presence of surface roughness. It is found that, in addition to the potential energy and the surface profile, another statistic parameter, called sampling number, also contributes significantly to the interaction between two rough surfaces. The two frequently observed facts that the nominally larger objects show weaker adhesion than smaller ones, and that the interaction function should be modified properly according to the resolution in modeling, can be considered as a purely statistic performance of the length scale effect at two limit cases, and can be further unified in our formula by the sampling number. Finally, the validation of our formula is discussed. Although it is not accurate for the JKR case, our simple formula provides a promising approach for predicting the adhesion especially for the DMT case and gives deep insight on the statistic nature in contact mechanics.

See the [supplementary material](#) for the detailed data shown in Fig. 4.

This work was supported financially by The Hong Kong Polytechnic University (Project 1-ZVQM).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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