Nanotribology investigations with classical molecular dynamics
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Chapter 2 *

Single asperity nanocontacts: comparison between molecular dynamics simulations and continuum mechanics models

Using classical molecular dynamics, atomic scale simulations of normal contact between a nominally flat substrate and different atomistic and non-atomistic spherical particles were performed to investigate the applicability of classical contact theories at the nanoscale, and further elucidate the phenomena that govern the perceived breakdown of continuum in nanocontacts. The Hertz elastic contact model was shown to sufficiently capture the results of molecular dynamics simulations prior to the onset of plastic deformation for non-adhesive contacts. Moreover, the results show that the slipping of dislocations was the deformation mechanism responsible for force drops during loading; prior to slipping, however, the plastic deformation was identified to be reversible. For adhesive contacts, the corresponding surface energies were calculated via molecular dynamics simulations and validated with values reported in the literature. The contact force and radius, as well as the contact stress distribution and dislocation propagation were then investigated in detail. Moreover, the results were compared to the limiting cases of the Johnson-Kendal-Roberts and Derjaguin-Muller-Toporov continuum models. The atomistic systems showed deviations from the classical models, which could be related to energy loss and changes in the effective work of adhesion, as well as the anisotropic properties of the atomistic systems. The findings of this chapter support the published literature on the subject, and further contribute to our understanding of discrepancies between atomistic and continuum descriptions of contact.

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Chapter 2: Single asperity nanocontacts

2.1 Introduction

Nanotribology is an interdisciplinary field of research that studies the friction, adhesion, lubrication and wear of nanocontacts. At the nanoscale, surface forces become dominant and they can influence the functionality of micro- and nano-sized devices. Similarly, contact is initiated at the atomic scale and is particularly important in modern technologies such as micro- and nano-electro mechanical devices, atomic force microscopy, and magnetic storage devices. Therefore, studying nanocontacts is a key step towards understanding the multiscale properties of contact phenomena.

In general, the structure of surfaces is random at the atomic scale with roughness occurring even for highly smooth surfaces. In order to isolate surface roughness from other physical parameters that affect tribology, many studies have focused on the contact of individual asperities with perfectly smooth surfaces [13]. A reason why single asperity studies have been so widely used is that deformations at the contacting interface can be described by continuum mechanics models [4]. In the present chapter, the focus is on classical single asperity studies to investigate whether these remain valid at the atomic scale. More specifically, the results were compared with the limiting cases of Johnson-Kendall-Roberts (JKR) [14] and Derjaguin-Muller-Toporov (DMT) [15] models.

Through the last decades, many researchers have applied theories of continuum contact mechanics to nanotribology measurements in order to determine fundamental parameters and processes at play in nanoscale contacts via experiments [16-19] and simulations [20-22]. Luan and Robbins [3, 23] examined nanocontacts via molecular dynamics (MD) simulations for different contacting tip geometries. They showed that there are deviations between atomistic behavior and continuum theory. In summary, they discussed that any realistic interactions lead to a gradual increase in repulsion with separation rather than an idealized hard-wall interaction. Also, surface roughness, which always exists in any discrete atomistic surface, results in pressure distributions on the surface that deviate from those described in continuum theory. Moreover, roughness has an influence on the work of adhesion. Besides these deviations, they also showed that friction theories do not hold at the nanoscale and, therefore, concluded that continuum models cannot correctly capture the behavior of nanocontacts. Later, Mo et al. [4, 5] showed that the break-down of single-asperity theories of friction depends on the method with which the contact area is measured. In continuum mechanics, the contact area is defined by the edge of the contact zone, or the area of the
asperity $A_{asp}$. However, because of the roughness at the atomic scale, the real contact area is different than $A_{asp}$. They defined the real contact as:

$$A_{\text{real}} = N_{ca} A_a,$$

where $N_{ca}$ is the number of contacting atoms defined by the atomic distances between the substrate and the tip, and $A_a$ being the average surface area per atom [4, 5]. The conditions in which continuum models break down are still under investigation, and this chapter aims to add to the study of the correspondence between continuum theory and atomistic behavior.

This chapter presents the results of the contact between different single spherical asperities and fcc-nickel substrates.

### 2.2 A short review of elastic contact theories

The first model for non-adhesive elastic contacts was published by Hertz [24] (e.g., see [25] for a review). This model describes the contact behavior of two contacting parabola, resulting in an elliptical contact area. For a simpler case of contact between a sphere of radius $R$ and a half-space penetrated by a depth $d_p$, which forms a circular contact area of radius $r$, the Hertz model is still valid. Assuming $d \ll R$, the following holds: $r^2 = Rd$. Hertz derived the pressure distribution of the form $p_{(l)} = p_0(1 - (l/r)^2)^{1/2}$, where $p_0$ is the maximum compressive pressure, with $l$ being the radial distance between 0 (at the center the contact) and $r$ (the contacting radius). Figure 2.1 (a) schematically illustrates a deformed sphere and pressure distribution for the Hertzian solution. The contact area radius $r$ can be obtained as a function of normal load $F_\perp$:

$$r^3 = \frac{3RF_\perp}{4 E^*},$$

with

$$E^* = \left(\frac{1 - \nu_1^2}{E_1} + \frac{1 - \nu_2^2}{E_2}\right)^{-1},$$

where $E_1$ and $E_2$ are the elastic moduli, and $\nu_1$ and $\nu_2$ are the Poisson’s ratios associated with each body. $E^*$ is known as the reduced Young’s modulus or contact elastic modulus. This model has been extensively studied in Chapter 4.

In 1971, Johnson et al. [14] proposed a new theory (JKR) that accounted for adhesion between the contacting bodies. This work was motivated by
experimentally measured contact areas that were larger than the areas predicted by Hertzian theory at low loads, and by the observation of a finite contact area at zero applied loads. In this approach, it was suggested that a tensile pressure distribution can be maintained at the contact area at zero separation, while neglecting the adhesion force outside the contact circle. They then proposed a pressure distribution of $\dot{p}(l) = -\dot{p}_0 (1 - (l/r)^2)^{-1/2}$, corresponding to the attractive adhesive force. This term was added to the pressure $p(l)$ given by Hertzian theory. The deformation and pressure distribution of an elastic sphere in contact with a rigid plane are shown in Figure 2.1 (b), for the JKR solution. Johnson et al. [14] derived the contact radius as:

$$r^3 = \frac{3}{4} \left( \frac{R}{E^*} \right) \left( F_\perp + 3\pi R \Gamma + \sqrt{6\pi R F_\perp + (3\pi R \Gamma)^2} \right),$$

(2.4)

where $\Gamma$ is the work of adhesion. This theory predicts a maximum tensile force of $-\frac{3}{2} \pi R \Gamma$, known as the pull-off force ($F_{po}$), necessary to detach the bodies.

Figure 2.1 Surface traction and the pressure distribution of an elastically deformed sphere regarding (a) Hertz and DMT, and (b) JKR solutions. The dotted blue lines (above the flat surface) show the compression, and the dashed red lines (under the flat surface) show the tension. The tension lines in (a) are based on DMT solution.

Subsequently, Derjaguin et al. [15] formulated their theory (DMT) to incorporate the effect of adhesion in contact by introducing the attractive forces outside the contact area (Figure 2.1 (a)). They showed that the deformed contact profile remained the same as in Hertzian theory, but with an overall higher load due to the adhesion force ($F_{ad}$) acting as a dead weight. They showed that the pull-off force is equal to $-2\pi R \Gamma$, and is reached at zero contact radius; the same conclusion was reached with the Bradley model for
rigid contacts [26]. Also, they found that the adhesive force gradually decreases to $-\pi R \Gamma$ as a function of displacement. Muller et al. [27] later showed that the contact radius under a zero load can be calculated for the DMT model by:

$$r_0^3 = \frac{3}{2} \frac{\pi R^2 \Gamma}{E^*},$$  \hspace{1cm} (2.5)

where $r_0$ is the contact radius at $F_\perp = 0$. Assuming constant adhesive force ($F_{ad} = F_{po}$), Maugis [28] showed that the DMT theory can be estimated by:

$$r^3 = \frac{3}{4} \frac{R}{E^*} (F_\perp + 2 \pi R \Gamma).$$  \hspace{1cm} (2.6)

![Figure 2.2](image)

**Figure 2.2** The contact radius vs load curves, for Hertz, DMT and JKR models, plotted by setting $E^* = 3/4$ GPa, $R = 1$ nm, and $\Gamma = \pi^{-1}$ N/m.

Figure 2.2 illustrates the different values of the contact radius between a spherical asperity and a flat surface with the Hertz, JKR and DMT solutions. In 1976, Tabor [29] analyzed both adhesive models, and suggested the following relation for the pull-off force:

$$F_{po} = -3/2 \pi R \Gamma \phi(\mu),$$  \hspace{1cm} (2.7)

where $\phi$ is a function of $\mu = \left(\frac{R \Gamma^2}{E^* r_E^3}\right)^{1/3}$, with $r_E$ being the equilibrium interatomic distance. The variable $\mu$, thereafter known as the Tabor parameter, is the ratio of the neck height formed in the JKR model over the equilibrium interatomic distance. Higher values of $\mu$ imply the applicability of
the JKR model, while the DMT model holds for small values of $\mu$. After some debate [30-33], Muller et al. [34] concluded that the two theories are limiting cases of a more general approach. It is known that the function $\phi(\mu)$ changes monotonically in the range between 1 (for the JKR limit) and $4/3$ (for the DMT limit); however, the function itself is not rigorously defined. It is possible to rewrite (2.7) as $\chi = - F_{po}/\pi R \Gamma$, and for the DMT and JKR limits, $\chi$ would be 2 and 1.5, respectively. Muller et al. [27] used the Lennard-Jones potential [35] to formulate the first elastic solution of adhesive contact (MYD) for a continuous variation of $\mu$ between the limits of the JKR and DMT models via a numerical method. Maugis [28] was the first to model elastic adhesive contacts for the whole range between the JKR and DMT limits.

All the above theoretical models deal with the contact of two homogenous, isotropic, axisymmetric contacting bodies. In addition, the solutions are driven by the following simplifying assumptions [25]:

a- surfaces are continuous and perfectly smooth,

b- strains are small enough for linear elasticity to be valid, and

c- surfaces are frictionless.

It should be noted that any deviation from these assumptions has an impact on the correlation between the models and experiments. For instance, for general anisotropic materials, there is no straightforward formula to estimate the value of the reduced Young’s modulus which is an essential part of continuum contact theories [36].

2.3 Simulation methodology

2.3.1 Overview of numerical experiments

A series of classical molecular dynamics (MD) simulations were utilized to study the nanocontacts between spherical single asperities and an atomically flat substrate, performed using the large-scale atomic/molecular massively parallel simulator (LAMMPS) [37]. The equations of motion were numerically solved using the velocity-Verlet algorithm [38] with an integration step size of 10 fs. This was selected from NVE-ensemble pre-test simulations as the largest time step that did not exhibit energy drift. Using the Berendsen thermostat algorithm [39], the temperature was kept constant at 300 K. Also, the [100], [010], and [001] crystallographic directions were selected as the $x$, $y$, and $z$ axes for the generation of all atomistic bodies. Moreover, the visualizations, dislocation analyses, and other atomistic related post processing steps were
done by the open visualization tool (OVITO) [40], and the crystal analysis tool (CAT) [41-44]. The various simulations performed in this study are summarized in Table 2.1.

The systems were generated from fcc-nickel and bcc-iron atoms. The integrated embedded-atom method potential database formulated and developed by Zhou et al. [45] was used for calculating the potential energy of the system. The elastic constants of the potential energies at 300 K were calculated via LAMMPS, and used for estimating the values of Young's modulus and Poisson ratio via Hill's approximation [46]. The results are summarized in Table 2.2.

Two categories of simulations were performed: one called forced particles (see section 2.3.2), and the other called free particles (see section 2.3.3). The systems with forced particles were simulated to extract the force-displacement and force-contact area data, while the systems with free particles were simulated to measure the contact area under zero externally applied force. All particles had a radius of 30 Å. The particles were made by cutting a sphere from a crystal that was aligned with the substrate, resulting in a terraced geometry. This geometry can be seen in Figure 2.3, which shows the “forced particles” described in more detail below. This kind of stepped atomic structure for nanoparticles has a good correspondence to experiments reported in the literature [47]. It should be mentioned that a slight tilt in the steps could result in a very different pressure distribution [23].

### 2.3.2 Forcéd particles

Three different single asperities were simulated: a non-atomistic repulsive asperity, and two atomistic ones with different crystal structures. In the non-atomistic case, the asperity was placed adjacent to the top of the substrate. However, for the atomistic asperities, the clearance between each asperity and the substrate was slightly larger than the potential’s cutoff radius. After equilibration, the particles were moved along [001̅] toward the substrate in a displacement control mode. The movement for the non-atomistic asperity was 10 Å from its center with a velocity of 1 m/s. In the case of atomistic asperities, the rigid layer was moved for 15 Å from its initial position with a velocity of 10 m/s; the latter was imposed due to the processing time limitations of MD. Then, the particles moved upward until the force between the particle and the substrate became zero. There was no relaxation time during or between the loading and unloading processes.
Table 2.1 A schematic summary of the numerical experiments, and their placement in the chapter:

<table>
<thead>
<tr>
<th>Non-atomistic indenter:</th>
<th>2.5.1 Contact with no adhesion</th>
</tr>
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<tbody>
<tr>
<td>contact with no adhesion</td>
<td></td>
</tr>
</tbody>
</table>

Surface energy: for both Ni and Fe along the \{100\} planes.

<table>
<thead>
<tr>
<th>Phase A:</th>
<th>2.5.2 Contacts with adhesion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phase B:</td>
<td></td>
</tr>
</tbody>
</table>

Free particle: for both Ni and Fe.

<table>
<thead>
<tr>
<th>2.5.2.1 Contacts of free particles</th>
</tr>
</thead>
</table>

Forced particle: for both Ni and Fe

| 2.5.2.2 Contacts of forced particles: Iron |
| 2.5.2.3 Contacts of forced particles: Nickel |

Table 2.2 The calculated elastic properties for nickel and iron for the used EAM potentials [45]. All values but the Poisson ratios are reported in the scale of GPa.

<table>
<thead>
<tr>
<th></th>
<th>( C_{11} )</th>
<th>( C_{12} )</th>
<th>( C_{14} )</th>
<th>( E )</th>
<th>( \nu )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>216.7</td>
<td>145.1</td>
<td>117.9</td>
<td>218.1</td>
<td>0.30</td>
</tr>
<tr>
<td>Fe</td>
<td>221.2</td>
<td>146.6</td>
<td>104.1</td>
<td>182.6</td>
<td>0.32</td>
</tr>
</tbody>
</table>
The geometric non-atomistic asperity was modeled as a purely repulsive soft (in reference to its force field) sphere. The interaction potential between the asperity and the substrate atoms was limited to distances $r \leq R$, where $r$ is the distance from the atom to the center of the asperity, and $R$ is the radius of the asperity. The geometrical asperity exerts a force of magnitude $-K(r - R)^2$ [48], where $K$ is a force constant related to the effective stiffness of the asperity that was set to 5 eV $\cdot$ Å$^{-3}$ [49].

The other two asperities were generated from atomistic nickel and iron. Each asperity was divided into two regions: the highest two atomic layers were laterally fixed and were only allowed to move along the z axis, thereby controlling the movement of the asperities. Below this section, a cylinder with a height of four atomic layers connected to a hemisphere was filled with non-constrained atoms. Figure 2.3 illustrates the geometry and atomistic representation of these two asperities.

![Figure 2.3 Atomistic representation of the (a) fcc-nickel, and (b) bcc-iron particles.](image)

### 2.3.3 Free particles

Using the continuum models, it is possible to predict the contact area between two bodies at a zero external load. To do this, two spherical particles were generated from nickel and bcc-iron and were allowed to come into contact with the substrate under zero external force. The atoms were not constrained, and each particle was placed 2 Å above the substrate. Since the EAM potentials’ cutoff radii were $\sim$6 Å, the particles were within the range of interatomic interactions with the substrate. The simulations were run for 1 ns
with zero applied external force. The results are discussed in section 5.2.2, and used through the discussion of forced adhesive particles.

### 2.3.4 Substrates

Different nanocontacts were simulated with various single asperity representations, and using two different substrates comprised of a single fcc crystal generated from nickel atoms. The substrates have dimensions of $40a_0 \times 40a_0 \times 20a_0$ and $40a_0 \times 40a_0 \times 10a_0$, for forced and free particles, respectively, where $a_0 = 3.52$ Å is the lattice constant for nickel. The size was validated through pretest simulations as being large enough to obtain reliable results; specifically, it was verified that the resulting stress fields were fully enclosed within the substrate material volume. Periodic boundary conditions (PBCs) were applied in the lateral directions. At the bottom, the last two atomic layers were fixed, resembling a rigid substrate. The next four atomic layers were assigned to be the thermostatic layer, and the remaining atoms were not constrained.

### 2.3.5 Evaluation of work of adhesion

The work of adhesion $\Gamma$ is the energy change of bringing a unit area of a phase $A$ into contact with a unit area of a phase $B$ in vacuum [50]. These two phases may be the same or correspond to different materials. By definition, $
abla = \gamma_A + \gamma_B - \gamma_{AB}$, where $\gamma_A$ and $\gamma_B$ are the surface energies of phases $A$ and $B$, respectively, and $\gamma_{AB}$ is the energy of their interface. If the phases are the same, then $\gamma_{AB} = 0$.

Continuum models require a value for $\Gamma$. This value can be calculated via MD simulations by subtracting the energy per unit area of the two atomically flat substrates in equilibrium with zero applied force (phase A) from the energy per unit area of the two substrates at infinite separation (phase B). To do so, a set of two simulations is needed [51]. In the first, two cubic atomic blocks, each with a size of $10a_0 \times 10a_0 \times 10a_0$, were constructed side by side. In the second simulation, there was a gap of $10a_0$ between the two blocks. The PBCs were applied in lateral directions. Each simulation was run for 150 ps. It should be noted that in these simulations, (001) surfaces formed both the interface and the generated free surfaces.

The above simulations were done for nickel and iron systems. However, in order to construct a system for a nickel/iron pair in the defined coordinate
system, the lateral lengths of the simulation box should be equal to the length of 287 unit cells of nickel and 352 unit cells of iron to ensure that the two are equal\(^\dagger\); otherwise, the PBCs cannot be applied in the lateral directions without introducing crystallographic defects to the system. Instead of running such a huge simulation, it is possible to estimate the value of \(\gamma_{AB}\) from \((\sqrt{\gamma_A} - \sqrt{\gamma_B})^2\) [50]. Therefore, in general, the work of adhesion can be expressed as \(\Gamma = 2\sqrt{\gamma_A\gamma_B}\). If \(A = B\), then \(\Gamma = 2|\gamma_A|\), where \(|\gamma_A|\) is the absolute value of the surface energy.

### 2.4 Contacting atoms and contact area

Mo et al. [4, 5] showed that the calculation of the real contact area is important in the study of contacts at the atomic scale. However, the calculation of the contact area is very challenging at this scale. The difficulty comes from various reasons, the most important of which are the detection of contacting atoms, and the definition of the area of contacting atoms.

Assuming that atoms can be represented as rigid spheres, their radii can be defined based on their crystallographic packing factors. There are 4 and 2 atoms in each fcc and bcc unit cell with packing factors of 0.74 and 0.68, respectively. Therefore, the atomic volume \((V_a)\) in these two crystal structures can be defined as \(0.74 \times a_0^3/4\) and \(0.68 \times a_0^3/2\) for fcc and bcc, respectively.

The projected atomic real area then can be defined as \(A_a = (3\sqrt{\pi}/4 V_a)^{2/3}\). Interaction forces between neighboring atoms vary with distance. In this work, a contact distance \(d_c\) was used to define atoms that were in contact; that is \(d_c = a_0/2 + r_a\), where \(r_a = (3/(4\pi) V_a)^{1/3}\) is the atomic radius. For the system with nickel and iron atoms, the mean value of the contact distances of nickel and iron was adopted as the contact distance between those different atoms. Hence, two atoms whose distance was equal to or smaller than the corresponding were defined as being in contact. The average of the contact areas of the contacting particle and substrate was reported as the contact area.

\[^\dagger\] The number of necessary unit cells can be found from \(N_{Ni} a_{0,Ni} = N_{Fe} a_{0,Fe}\). Furthermore, it should be noted that \(N\) should be an integer, and not a fraction.
2.5 Results and discussion

2.5.1 Contact with no adhesion

The force-displacement (\(F - d\)) curves of the contact of the non-atomistic asperity are shown in Figure 2.4. The curve shows a sequence of drops that are directly related to plastic deformations. The first drop occurred at a depth of \(~4.8 \text{ Å}~\), and is an indication of the onset of plasticity. At this point, the elastic stress under the indenter was partially relieved by plastic deformation.

![Figure 2.4](image)

**Figure 2.4** The force-displacement curve of the non-atomistic repulsive contact. For the Hertz solutions, \(E^* = 239.7 \text{ GPa}\) and \(E^* = 244.8 \text{ GPa}\) were obtained from the calculated elastic properties and fitting the Hertz formula to the simulation results, respectively.

In order to confirm that the deformation before the indentation depth of \(4.8 \text{ Å}~\) was reversible, the crystal structure of the system was analyzed via an additional simulation. In this simulation, the non-atomistic particle indented the substrate to a depth of \(4.5 \text{ Å}~\), and was then returned to its original position. All other parameters of the system were the same as before. Figure 2.5 shows the dislocation generations during this process. It is found that, at a depth of...
3.5 Å, the nucleation of dislocations was started, and the process was continued to a depth of 4.5 Å. These generated dislocations were mainly Shockley partials with Burgers vectors \( \mathbf{b} = (a_0/6)[\bar{2}11] \) and \( \mathbf{b} = (a_0/6)[2\bar{1}1] \). In two points, these dislocations interacted with each other as follows:

\[
\frac{a_0}{6} [\bar{2}11] + \frac{a_0}{6} [2\bar{1}1] = \frac{a_0}{3} [01\bar{1}] .
\]

(2.8)

The newly generated dislocations are stair-rod sessile, known as Lomer-Cottrel locks [52] with their Burgers vectors on, both, (111) and (\( \bar{1}11 \)) planes. These locks act as strong barriers; however, their disappearance in further indentations can be explained by the unzipping mechanism [53]. As the next step, the indenter was moved to its initial position, in which all dislocations were removed from the crystal structure at an indentation depth of 2.5 Å, as shown in Figure 2.5 (h). By analyzing the crystal structure at the end of the unloading process, it was found that the system returned to a perfect crystal structure; a side view in Figure 2.5 would only show surface atoms. This process indicated that the performed plastic deformation was reversible [54].

\[\text{Figure 2.5} \text{ Dislocation evolutions during (a-d) the loading and (e-h) unloading processes. The green dislocations are Shockley partials, and the red ones are stair-rods. The surface of the deformed part including undefined defects is also shown.} \]

\[\text{The atoms are colored based on their centrosymmetry parameter values.} \]

In fcc crystal structure, the smallest Burgers vector has a length of \( a_0/\sqrt{2} \). In Figure 2.6, the displacement vectors of the atoms, whose magnitudes were larger than the smallest Burgers vector, are depicted at four different indenter
depths of 4.5 Å and 5.0 Å, before and after the onset of plastic deformation, respectively, and at 9.5 Å and 10.0 Å, representing the final snapshots of the loading process. As can be seen, during the reversible regime of deformation, the atomic movements were restricted to those atoms that were adjacent to the indenter. By the initiation of plastic deformation, the slipping process was activated mainly in (1̅11)[1̅0̅1] and (1̅1̅1)[0̅1̅1] slip systems. Figure 2.6 shows that the process was continued until a depth of 9.5 Å; however, at the depth of 10.0 Å the movement of the atoms reached the edges of the simulation box and the simulation, at least for purposes of mechanical analysis, was no longer valid. The surface profile of the indentation at the depth of 9.5 Å, and its corresponding von-Mises stress field are shown in Figure 2.7. The edges of the pile-up profile align with [1̅10], [̅1̅0], and [100].

![Figure 2.6](image)

**Figure 2.6** The displacement vectors (colored yellow) of the atoms which their movement were larger than the smallest Burgers vector, at different indentation depths. The figures are shows the system from (upper row) side view and (lower row) top view.

Because in fcc crystal structures the main slipping plane is \{111\}, the substrate was sliced along one of these planes, and the von-Mises stress field during the simulation was extracted as illustrated in Figure 2.8. As soon as the plastic deformation was initiated, the shape of the stress field was changed slightly, and a considerable amount of local stress was released at the depth of 5.5 Å due to dislocation slips, as shown in the figure. From the top view, a stress field becomes visible at the corners of the simulation box. This field is the result of slipping dislocations in the system.
Results and discussion

Figure 2.7 (a) Surface profile, and (b) surface stress distribution of the substrate at an indentation depth of 9.5 Å.

Figure 2.8 The von Mises stress field during the loading process along a \(\{111\}\) plane. The fields are depicted from an atomic slice of the substrate beneath the indenter.

The movement of the dislocations at the depths of 5.0 Å, and 5.5 Å is depicted in Figure 2.9. Moreover, it should be noted that the stress field of the system is higher along the slipping planes, as can be seen in Figure 2.10.

The reversible part of the \(F - d\) curves is well described by the power law relation \(F = Cd^m\), where \(C\) and \(m\) are fitting constants. The value of \(m\) was found to be 1.23, which is close to 1.5, the value reported for spherical indenters [55]. This force estimation of the elastic deformation part was also plotted in Figure 2.4, and shows a very good correlation to the simulation results.
Because the particle had no adhesive force, the Hertzian formulation is expected to apply when modelling the elastic deformation region. In this model, the normal applied force can be calculated by:

$$F_\perp = \frac{4}{3} E^* R^{0.5} d^{1.5}. \quad (2.9)$$

Assuming the particle to be a rigid body, its Young’s modulus would be infinity; in other words, the value of the reduced Young’s modulus would be only a function of the mechanical properties of the substrate, i.e. $E = 218.1$ GPa and $\nu = 0.3$ (see Table 2.2). The elastic contact modulus, by means of (2.3), would be calculated to be $E^* = 239.7$ GPa. By fitting the Hertz solution to the MD results, the value of the reduced Young’s modulus was found to be 244.8 GPa, which barely changes the Hertzian solution, as plotted in Figure 2.4. The slight discrepancy between the Hertzian solution and MD results could be partly due to the fact that, although the deformation was reversible, it was not purely elastic. It should be noted that higher values of the force constant of the particle would increase the reduced Young’s modulus [56]. Also, increasing the indenter’s radius results in higher values of $E^*$ [57].
Moreover, the temperature of the substrate, and the moving velocity of the particle might have their own influence on the results; however, this is not the focus of the current investigation.

### 2.5.2 Contacts with adhesion

Figure 2.11 demonstrates the contacts at the beginning of the simulation, at their highest compression pressure, and after detachment, for both forced particles. The metal transfer in the system with the iron particle was small. On the other hand, the nickel particle had a very significant amount of metal transfer to the surface of the substrate. Such behavior was expected [21], and indicates high adhesion between the two surfaces. Although it is clear that the system with the nickel particle exhibited ductile rupture, unlike the detachment in the system with the iron particle, it was examined whether the contact behavior of the systems prior to sever plastic deformations can be described by continuum contact theories.

![Figure 2.11](image)

**Figure 2.11** The sliced side view of the atomistic configurations of (a) nickel and (b) iron forced particles contacts with the substrate.
As the first step, the work of adhesion for each contacting systems was calculated. The potential energies of the systems when the blocks were side by side (phase A), and when they were far from each other (phase B) are reported in Table 2.3. In order to calculate the real contact area of the interface, the number of contacting atoms should be calculated first. Using the (001) plane density for fcc \((2/a_0^2)\) and bcc \((1/a_0^2)\) structures, it can be found that in an area of \(10a_0 \times 10a_0\) with a (001) plane, there are 200 and 100 atoms in fcc and bcc crystal structures, respectively. It should be mentioned that the same results can be obtained also by using the corresponding contact distances for the calculation of the number of contacting atoms. The values of \(A_{\text{real}} = N_{ca}A_a\) were obtained, and the surface energies were calculated by dividing these two values as reported in Table 2.3. The calculated surface energies are close to experimental values reported by other researchers [58].

Considering the two kinds of adhesive contacts in this study, the values of the work of adhesion would be as follows: for the pure nickel system, the work of adhesion could be defined as \(\Gamma_{\text{Ni/Ni}} = 2|\gamma_{\text{Ni}}| = 4.32\) N/m; similarly, the nickel/bcc-iron contact would have \(\Gamma_{\text{Fe/Ni}} = 2\sqrt{\gamma_{\text{Fe}}\gamma_{\text{Ni}}} = 4.54\) N/m.

<table>
<thead>
<tr>
<th></th>
<th>Energy A</th>
<th>Energy B</th>
<th>(A_{\text{real}}) (Å²)</th>
<th>(\gamma_{(100)} = \Delta\text{Energy} / \text{Area})</th>
<th>(\gamma_{\text{exp}} [58])</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni/Ni</td>
<td>-35858.8</td>
<td>-35569.6</td>
<td>2144</td>
<td>-2.16</td>
<td>-2.45</td>
</tr>
<tr>
<td>Fe/Fe</td>
<td>-17245.6</td>
<td>-17071.9</td>
<td>1164</td>
<td>-2.39</td>
<td>-2.48</td>
</tr>
</tbody>
</table>

### 2.5.2.1 Contacts of free particles

The free particles were attracted toward the substrate because of the interatomic forces. Figure 2.12 shows the systems at the end of the simulations. The nickel particle was deformed at the contact, while the iron particle largely preserved its initial shape. Both systems were analyzed for crystallographic defects. It was found that no dislocation was generated in the substrate in the system with the nickel particle, while the particle itself experienced plastic deformation via generation and sliding of dislocations.
These observations agree with experimental investigations that showed that plastic deformation may occur during adhesive interaction even with no external load [16]. In the system with the iron particle, it was only the substrate that experienced a limited number of dislocation generations: there were two Shockley partials, which stopped as soon as they interacted, and a stair-rod partial generated in the system.

The contact areas were calculated by measuring the number of contacting atoms as summarized in Table 2.4. Assuming the contact areas as perfect circles, the values of contact radii were obtained to be $r_{0,Ni/Ni} = 2.09$ nm, and $r_{0,Fe/Ni} = 1.73$ nm. These results are compared to the corresponding values of JKR and DMT models in Table 2.5. The comparison shows that both of the models underestimate the nickel/nickel contact area; however, the contact area of iron/nickel was correctly described by the JKR model.

Table 2.4 The number of contacting atoms and area of the free particles rested on the substrate.

<table>
<thead>
<tr>
<th></th>
<th>Particle atoms</th>
<th>Substrate atoms</th>
<th>Average contacting area (Å²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni/Ni</td>
<td>260</td>
<td>304</td>
<td>1371.5</td>
</tr>
<tr>
<td>Fe/Ni</td>
<td>199</td>
<td>187</td>
<td>937.5</td>
</tr>
</tbody>
</table>
Table 2.5 The contact modulus for the contacting systems, and the contact radius estimated from simulation and JKR and DMT models. The radii are reported in nm.

<table>
<thead>
<tr>
<th></th>
<th>$E^*$ (GPa)</th>
<th>$r_{0,\text{simulation}}$</th>
<th>$r_{0,\text{JKR}}$</th>
<th>$r_{0,\text{DMT}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni/Ni</td>
<td>119.8</td>
<td>2.09</td>
<td>1.66</td>
<td>1.15</td>
</tr>
<tr>
<td>Fe/Ni</td>
<td>110.0</td>
<td>1.73</td>
<td>1.74</td>
<td>1.21</td>
</tr>
</tbody>
</table>

2.5.2.2 Contacts of forced particles: Iron

Figure 2.13 shows the force-displacement ($F - d$) curve of the iron/nickel system. The force drops at the very beginning corresponding to pull-in adhesion. This pull-in force was found to be $F_{\text{pi}} \approx -35 \text{ nN}$. The drops in the adhesion force are stepped because of the terraced geometry of the particle. Then, the force increases smoothly up to its maximum value with no sign of plastic deformation. As the unloading starts, the force drops suddenly to its minimum value. Although the particles did not experience a sudden pull-off in this simulation, the minimum force was referred to as the pull-off force in this chapter. The pull-off force was found to be $F_{\text{po}} \approx -105 \text{ nN}$. Then, the force increases up to the point where its slope starts to change. This change relates to plastic deformations in the system. As the unloading process continues, the particle separates and the force jumps to zero.

![Figure 2.13](image-url)
The analysis of the atomic configuration shows that there was no dislocation in the system during the unloading process until the force reached its minimum (point $F_{po}$ in Figure 2.13). Following that point, however, the particle experienced plastic deformation. Figure 2.14 shows the deformed particle and the dislocation lines in it at the end of the simulation. The entire lower part of the particle was deformed, and experienced deformation twinning along the $(112)$ plane to obtain the coherent twin. The angle between the twin-surface intersection and the fixed layer of the particle was found to be $\sim 32^\circ$. This value is very close to its ideal angle, i.e. $\theta \approx 35^\circ$ (e.g., see [59] for a review).

**Figure 2.14** Atomistic configurations of the deformed bcc-iron forced particle at the end of the simulation.

Considering that JKR and DMT models do not describe the hysteresis of the $F - d$ curves, both pull-in and pull-off forces were used for calculation of $\chi$. The values of $\chi$, which are reported in Table 2.6, were found to be out of the JKR-DMT limits: $\chi(F=F_{pl}) = 0.82$ was lower than $\chi_{JKR} = 1.5$, and $\chi(F=F_{pl}) = 2.45$ was higher than $\chi_{DMT} = 2$.

**Table 2.6** The pull-in and pull-off forces, and the corresponding values of $\chi = -F/\pi R \Gamma$ for the iron/nickel forced particle system.

<table>
<thead>
<tr>
<th>$F_{pi}$ (nN)</th>
<th>$\chi(F=F_{pi})$</th>
<th>$F_{po}$ (nN)</th>
<th>$\chi(F=F_{po})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-35$</td>
<td>0.82</td>
<td>$-105$</td>
<td>2.45</td>
</tr>
</tbody>
</table>
The simulation shows that the contact area decreases until the particle is separated from the substrate; however, because of the transferred atoms between the particle and the substrate, the calculated contact area would be unacceptably overestimated if it were measured after the pull-off. Therefore, it was measured for the system during the loading and the unloading processes only up to the pull-off. The simulation results and the JKR and DMT solutions are shown in Figure 2.15. Two values of the contact area were obtained from the simulations: the real contact area $A_{\text{real}}$ based on (2.1), and the projection of the contacting atoms $A_{\text{pc}}$.

**Figure 2.15** Changes of the contact area as a function of the applied normal force in the system with the iron forced particle. The loading step begins from point A, and continues to its maximum load at point M. By the unloading step, the force reduces to its pull-off value at point P.

The inception of contact occurs at point A. Then, the contact area increases while the force drops because of the pull-in adhesion force. It can be seen that when the force reaches zero again, the contact area is very close to the JKR prediction, which was expected from the results of the free particle simulations. Moreover, as the loading increases the system’s behavior was very close to the JKR model, till the substrate start to generate dislocations. As a consequence, the simulated system’s behavior deviated from the JKR model. Then, the unloading step begins, but the contact area increases slightly; this might be a result of the kinetic energy of the particle’s atoms: i.e., after changing the direction of the force, atoms were still moving in their previous
Results and discussion

direction for a brief time. As the unloading continues, the force drops but the contact area remains almost unchanged until pull-off occurs. The comparison between $A_{\text{real}}$ and $A_{\text{pc}}$ shows that the $A_{\text{pc}}$ is not that sensitive to the generation of dislocations. Moreover, the analyses of the contact showed that the contacting atoms had a bowl shape, noticeable during the unloading process; while the 3D shape of the contact cannot be reflected in the 2D projection of the contacting atoms, the actual shape of the contact affects the real contact area. Therefore, as soon as the dislocations were generated, the values of $A_{\text{pc}}$ were smaller than of $A_{\text{real}}$. Moreover, it was found that, during the loading process, the results of the projection of the contacting atoms were closer to the JKR model than to $A_{\text{real}}$.

2.5.2.3 Contacts of forced particles: Nickel

The $F - d$ curve of the system is shown in Figure 2.16. At the beginning of the loading process the force drops to $F_{\text{pi}} \approx -22$ nN because of the adhesion force between the asperity and the subject. The adhesion-caused drops are shown to occur more than once; that is because of both the stepped geometry of the particle and the high value of the adhesion force. As the loading process was continued, there were abrupt force drops as indications of plastic deformations. By starting the unloading process, the force drops suddenly to a minimum of $-190$ nN. Then, the curve continues with a saw-like shape, indicating a number of plastic deformations, until the final rupture.

![Figure 2.16](image)

**Figure 2.16** The vertical force vs displacement curve of the bcc-iron forced particle systems with the displacements of 1.0 and 1.5 nm.
An additional MD simulation was run to avoid inducing plastic deformation during the loading process by reducing the displacement of the particle to 1.0 nm. The $F - d$ curve of this additional simulation is also shown in Figure 2.16. During the unloading process, the force reached its first local minimum, referred as the pull-off force in this chapter, with a value of $F_{po} \approx -67$ nN. The atomic configuration analysis of the system showed no dislocation line in the system at pull-off. As the unloading process was continued, instead of an abrupt detachment, the particle experienced a series of plastic deformations along \{111\} planes, which resulted in the saw-like shape of the curve.

The values of $\chi$ using, both, the pull-in and pull-off forces were calculated to be 0.54 and 1.65, respectively. The results (summarized in Table 2.7) indicates the applicability of the JKR model for describing the contact.

**Table 2.7** The pull-in and pull-off forces, and the corresponding values of $\chi = -F/\pi R \Gamma$ for the nickel/nickel forced particle system.

<table>
<thead>
<tr>
<th>$F_{pi}$ (nN)</th>
<th>$\chi(F=F_{pi})$</th>
<th>$F_{po}$ (nN)</th>
<th>$\chi(F=F_{po})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>-22</td>
<td>0.54</td>
<td>-67</td>
<td>1.65</td>
</tr>
</tbody>
</table>

The evolution of the contact area between the particle and the substrate was measured and is depicted in Figure 2.17. As the loading begins, the adhesion force becomes increasingly dominant at the interface. The contact area increases until the force reaches its maximum. For the force ranging between zero and its maximum (point M), the JKR model compares favorably as opposed to the DMT. As the unloading process starts, the contact area remains unchanged.

### 2.6 Conclusions

The results presented in this chapter address some discrepancies between the continuum models and atomistic simulations for the contact of simple geometries. For contacts with no adhesion, it was shown that Hertzian theory can describe the force-displacement behavior of the system for the reversible regime of deformation. Also, it was shown that the stress field of the substrate beneath the indenter in the slipping plane was similar to that predicted by continuum models; however, it should be noted that the stress field is
Figure 2.17 Changes of the contact area vs. the applied vertical force in the system with the nickel forced particle. Point A indicates the beginning of the simulation which goes up to point M, corresponding to the maximum load. The force drops to its pull-off value (point P) as the unloading starts.

directional in crystalline atomic systems, and therefore, not the same as what is assumed in continuum models.

Although the results were in general agreement with Hertzian theory, continuum adhesive models (JKR and DMT in this study) were unable to correctly describe atomic scale contacts when adhesion was introduced to the system. One of the difficulties in the comparison between MD simulations and continuum models was the elastic/plastic deformation of the particles after the systems reached pull-off; in comparison, continuum mechanics models predict that the particles suddenly detach from the substrate at pull-off. It was also shown that both systems experienced some plastic deformation after pull-off. The iron particle detached from the substrate and exhibited twinning behavior. On the other hand, the nickel particle formed extensive necking at the interface with the substrate, and experienced ductile rupture instead of detachment.

Analyzing the free particles, it was found that the JKR model could correctly predict the contact area of the iron particle; however, both, JKR and DMT models underestimated the contact area of the nickel free particle. Considering the plastic deformations of the nickel free particle during the process, this discrepancy could be justified.

Comparing the contact area of the forced particles as a function of force, it
was found that during the initial stage, i.e. pull-in and for $F_\perp \leq 0$ nN, the contact area increases suddenly, but in an stepped manner. This stepped behavior, which could also be recognized in the $F - d$ curves, was a direct consequence of the terraced geometry of the forced particles. While this geometry affected the contact’s behavior to deviate from continuum models, the loading contact could be correctly described using the JKR theory, as long as no dislocation line was generated in the system; the substrate experienced dislocation generation in the contact with the iron particle, which increased the contact area, and resulted in a deviation from the JKR model.

Although the JKR and DMT models are reversible, MD simulation results show an adhesion hysteresis for both atomistic asperities. This behavior is related to energy dissipation because of inelastic deformation in the contact, which affects the work of adhesion [60]. Considering the energy loss, the effective work of adhesion is reduced during the loading step; reducing the load necessary to separate the surfaces, in turn causes the effective work of adhesion to be increased. Therefore, the contact area remains unchanged until the force can overcome the increased effective work of adhesion.

Deviations from continuum models, as demonstrated in this chapter and as suggested in the literature [3, 23], are to be expected when studying atomistic systems since many fundamental assumptions of those models are neglected: for example, materials are not necessarily homogenous, and the contact surfaces are not frictionless. However, the results show that atomistic contacts, during the loading process, can be correctly described by the continuum models as long as the system has not experienced dislocation generation. The reason is that the generated dislocations introduce extra atomic steps at the contact’s interface, which results in an unpredictable increment in the contact area. It should be noted that the generation of dislocations at their early stage leads to reversible plastic deformation; therefore, the reversibility of the deformation is not enough for claiming the applicability of the continuum models.