Molecular dynamics nano-scratching of aluminium: a novel quantitative energy-based analysis method

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Abstract

Atomistic models for friction suffer from the severe length- and time-scale restrictions of molecular dynamics. Even when they yield good qualitative results, it is difficult to draw meaningful quantitative conclusions from them. In this paper, a novel approach to quantify the scratching work and the energy associated with the creation of plastic zones is presented. The approach is combined with a statistical criterion to determine the significance of simulation box size and sliding rate effects on the friction coefficient. These two methods are applied to a large parametric molecular dynamics study of single-crystal single-asperity aluminium nano-scratch with varying simulation sizes, indentation depths and scratching speeds in order to analyse these size and rate effects. The results show that the simulation size effects are a considerable obstacle to understanding the atomistic origins of friction – using present-day computing hardware – as they have a strong influence on the core mechanisms of sliding friction. A motivation for the development of a new 3D multi-scale method for a hybrid nano- and micro-scale description of plasticity is formulated.

Keywords: Molecular Dynamics, Dislocations, Friction, Plasticity

1. Introduction

Frictional contact is a much studied, yet poorly understood phenomenon with great impact on many everyday processes. Most laws describing friction have been developed for macro-scale contact [1, 2, 3]. Micro- and nano-scale friction phenomena occurring in micro- and nanoelectromechanical devices (MEMS and NEMS, respectively) such as hard drives and atomic force microscopes, are generally insufficiently described by such laws [4]. It has been shown that macro-scale modelling of contact breaks down at the nano-scale and in the case of single-crystal metal contact it is virtually always accompanied by plasticity [5].

This present study aims at an improved understanding and quantitative description of dry sliding contact between a single nano-scale asperity and single-crystal metal. This simple case is a key element in uncovering the atomic origins of friction in general and in quantifying the friction work and the energy associated with the creation of plastic zones.

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Several attempts to understand nano-scale contact at the atomic scale (see e.g., [6, 4, 7, 8, 9]) suffer from the size and time scale restrictions of molecular dynamics (MD). There have been many studies investigating effects of indenter shapes and sizes (e.g., [10, 11]), surface roughness (e.g., [12]), indentation depths and third bodies (e.g., [13]) but to the best of our knowledge, none have quantitatively investigated how the plastic mechanisms involved with friction depend on the size of simulated domains.

Larger scale discrete dislocation dynamics (DD) have also been used (see e.g., [14, 15]) to relax these size and time scale restrictions and to study contact at the micro-scale. Dislocation nucleation, however, is poorly understood and modelled using ad-hoc nucleation criteria which have an important impact on the results obtained from DD simulations, see [15]. Furthermore, to our knowledge, only two-dimensional discrete dislocation methods have been applied to contact, thus defying the inherently 3D nature of dislocation nucleation in indentation scenarios [16].

A large parametric study in which we scratch single crystal aluminium substrates of varying thickness with a spherical indenter at varying indentation depths and scratch speeds is performed. We then present a novel MD approach to quantify the energy associated with plastic deformation in nano-scale contact between crystalline metals. This approach is subsequently applied to the parametric study in order to determine the size and rate dependencies of scratching work in general and of the energy associated with plastic deformation. We also define a statistical criterion to determine whether there is a significant influence of size and rate effects on the friction coefficient.

The results from these analyses are used to detail the inherent problems with seeking to understand the atomistic origins of friction with single-scale models on present-day computing hardware. They also motivate the development of a new 3D multi-scale method for the modelling of micro-scale plasticity with an atomistic treatment of dislocation nucleation.

2. Parametric scratch study

We study aluminium substrates scratched by a rigid spherical aluminium indenter at low temperatures. Substrates of varying thicknesses $h$ are indented to varying depths $\Delta y$ and scratched at constant $\Delta y$ and scratching speed $v$, see Figure 1. For one $\Delta y - h$ combination, $v$ is varied as well. There are three mechanisms through which the scratching work $W_{sc}$ is absorbed by the substrate:

1. **Direct heat generation** $Q_1$
   The indenter induces lattice vibrations while scratching.

2. **Permanently stored deformation energy** $E_{pl}$
   The structure of the crystal beneath the indenter can become plastically deformed, e.g., through the creation of dislocation networks.

3. **Secondary heat generation** $Q_2$
   Dislocation motion radiates heat [17].

A novel method to separate the scratching work $W_{sc}$ into dissipated heat $Q = Q_1 + Q_2$ and permanently stored deformation energy, or plastic energy, $E_{pl}$ is presented and applied.

![Fig. 1. Schematic depiction of scratch parameters; $h$ is the substrate thickness, $\Delta y$ the indentation depth. The scratch speed $v$ is applied by imposing the indenter position $l(t) = vt$ at time $t$.](image)
2.1. MD Simulation

Setup. The scratching simulation is started at absolute zero temperature $T = 0\,\text{K}$ in MD. Figure 2(a) shows the general setup; the spherical indenter (dark red atoms) scratches over a single crystal aluminium substrate. Only half the system is shown to visualize deformation under the surface.

Both the indenter and the substrate are carved from the same crystal (and therefore perfectly commensurate), and the Miller indices of their crystallographic orientation are shown in Figure 2(b). Aluminium has a face centred cubic (FCC) lattice with a lattice constant of $a = 4.04\,\text{Å}$ if expressed in crystal directions $\langle 100 \rangle$. In the $(x, y, z)$ coordinate system, the lattice constants in each direction become

\begin{align*}
    a_x & = \left\| \bar{1}\bar{1}\bar{2} \right\| = \sqrt{6}a = 9.91\,\text{Å}, \\
    a_y & = \left\| \bar{1}\bar{1}0 \right\| = \sqrt{2}a = 5.72\,\text{Å}, \\
    a_z & = \left\| \bar{1}\bar{1}\bar{1} \right\| = \sqrt{3}a = 7.01\,\text{Å}.
\end{align*}

Indentation and scratch directions are $[1\bar{1}0]$ and $[1\bar{1}\bar{2}]$, respectively. The length and width of the substrate (in $x$ and $z$ directions) are kept constant at 64 lattices (634 Å) and 16 lattices (113 Å). The spherical indenter has a diameter of 46 Å.

Boundary conditions. The $x$ and $z$ directions have periodic boundary conditions, the two bottom layers of atoms (i.e., all atoms within $r_f = 2.9\,\text{Å}$ from the bottom boundary) are kept fixed (grey atoms in Figure 2(b)) and the indenter displacement is imposed. All other atoms are free to move and subject to the Mendeleev embedded atom model (EAM) potential for aluminium [18].

(a) Snapshot during simulation: dark red atoms belong to the rigid indenter. The other atoms are coloured according to their displacement (dark blue: no displacement). Only half of the simulated domain is shown in order to make the plastic deformation in the bulk under the indenter and in the groove behind the indenter visible.

(b) Schematic simulation setup: Grey atoms in the bottom layer represent fixed atoms within $r_f$ from the bottom boundary, $F(t)$ is the $x$ component of the force acting on the indenter, $r_i(t)$ and $\dot{r}_i(t)$ are the position and velocity vector of atom $i$. The crystal orientation of both the indenter and the substrate with respect to the coordinate system is shown in Miller indices.

Fig. 2. MD scratch setup with aluminium indenter and substrate
Parameter space. Next, we describe the range of parameters which has been explored. The varied parameters are

- the thickness \( h \):
  \[ h \in \{4, 8, 16, 32\} \text{ Å}, \]
- the indentation depth \( \Delta y \):
  \[ \Delta y \in \{0, 1, 2, 5, 10\} \text{ Å}, \]
  where \( \Delta y = 0 \) is the case of grazing contact,
- and the scratching speed \( v \):
  \[ v \in \{5, 10, 20, 40, 80\} \text{ m/s}. \]

Rather than exploring the entire parameter space, which would be computationally very costly, two sub-spaces have been explored:

- **Constant speed**
  for \( v = 10 \text{ m/s} \) all scratches for \( \{h\} \times \{\Delta y\} \) have been simulated in order to examine size dependence.
- **Constant thickness**
  for \( h = 22.9 \text{ Å} \) all scratches for \( \{v\} \times \{\Delta y\} \) have been simulated in order to examine rate dependence.

Simulation. All simulations are performed with LAMMPS [19] used through the multi-scale modelling framework LibMultiscale [20]. This combination has been chosen, because LibMultiscale allows the simple implementation of constrained minimisation problems for molecular statics (MS). For initialisation, the substrate is indented in \( y \) direction to the predefined indentation depth. The heat and mechanical waves generated by the indentation are then quenched out in a MS minimisation of the potential energy \( E_{\text{pot}} \) in order to start from a cold crystal. The minimisation is discussed in some detail below in Section 2.2. Once equilibrium is reached, the indenter is moved in negative \( x \) direction at constant \( v \). For later analysis, the forces acting between the indenter and the substrate are evaluated and periodic snapshots \( R(t_j) = R(\{r_i(t_j), \dot{r}_i(t_j)\}) \) of the state of the system are taken, where \( r_i(t_j) \) and \( \dot{r}_i(t_j) \) are the position and velocity of atom \( i \) at time \( t_j \), see Figure 2(b). The period \( t_{j+1} - t_j \) is chosen so that there is one snapshot per ångström of covered scratch distance.

2.2. Computation of permanently stored deformation energy

Energy balance. The energy influx into the system is due to the scratching work \( W_{\text{sc}} \)

\[
E_{\text{in}}(t) = W_{\text{sc}}(t) = \int_0^t F(\tau)v \, d\tau,
\]

and it is stored in the system\(^1\) in the form of kinetic and potential energy,

\[
E_{\text{tot}}(t) = E_{\text{kin}}(t) + E_{\text{pot}}(t).
\]

We are not directly interested in \( E_{\text{kin}} \), and \( E_{\text{pot}} \) is given by the Mendelev EAM potential of the form

\[
E_{\text{pot}}(t) = \frac{1}{2} \sum_{ij} V(r_{ij}) + \sum_i \Phi(\rho_i),
\]

\(^1\)Note that this energy balance is valid because all simulations presented here have been run without a thermostat. Therefore, the substrate heats up during the scratch. A subset of the explored parameter space, however, has been repeated with a velocity scaling thermostat layer [21] above the fixed bottom atoms in order to drain excess heat (not presented here). The measured forces with or without thermostat do not differ significantly so the decision has been made to take advantage of the exact energy balance \( (4) \).
where \( V(r_{ij}) \) is a pair potential, \( r_{ij} \) is the distance between atoms \( i \) and \( j \) and \( \Phi \) is the embedding energy required to insert an electron into the electron density \( \hat{\rho}_i = \sum_{j \neq i} \rho(r_{ij}) \) at the position of atom \( i \) due to all other atoms. The functions \( V, \Phi \) and \( \rho \) are semi-empirical potentials.

The energy influx equation (4) tells us that the energy of the system will increase during the scratch. The heat generated by the scratch is directly linked to an increase in both \( E_{\text{kin}} \) and \( E_{\text{pot}} \). However, the scratching causes atoms under the indenter to reorganise and leave a trace of permanent plastic deformation of the crystal. This plastic deformation defines a new – now imperfect – crystal structure and is also associated to a variation (usually an increase) of \( E_{\text{pot}} \). We call this variation \textit{permanently stored deformation energy} or short, \textit{plastic energy} \( E_{\text{pl}} \).

A way to determine which atoms participate in plastic deformation – and are therefore part of the plastic zone – is to compute their centrosymmetry parameter \( P \) which is zero for FCC material under homogeneous elastic deformation and nonzero for plastic deformation [22]. Atoms in an FCC lattice for which \( P > 0.5 \) are part of a plastic event which is at least a partial dislocation and we refer to them as \textit{dislocated atoms}.

Figure 3 shows four snapshots taken during a scratching simulation which clearly depict these changes in the crystal structure. Indenter atoms (blue), dislocated atoms (red) and atoms belonging to the chip and the groove (white) are represented as spheres. All other atoms are plotted as transparent white dots. We observe that there is a trail of plasticity behind the indenter, but also that the scratch causes dislocation loops to nucleate under the indenter (Figure 3(b)) and to travel into the bulk.

Determining \( E_{\text{pl}} \) quantitatively requires a method to split \( E_{\text{pot}} \) into a thermal and a structural component,

\[
E_{\text{pot}}(t) = E_{\text{pot,therm}}(t) + E_{\text{pot,struct}}(t).
\]

(7)

Keeping in mind that the simulation starts at 0 K and stays close to 0 K, thermal expansion of the crystal is negligible. Therefore, \( E_{\text{kin}} \) and \( E_{\text{pot,therm}} \) can be eliminated from the crystal by molecular statics (MS) minimisation.

An MS computation’s goal is to find the closest local minimum of \( E_{\text{pot}}(\{r_i\}) \) starting from a given snapshot \( \{r_i(t)\}^2 \). A local minimum of \( E_{\text{pot}} \) is a state \( \{r_i^{\text{min}}\} \) for which all atoms are at equilibrium

\[
\{\{r_i^{\text{min}}\} = \arg \min_{\{r_i\}_{\text{free}}} E_{\text{pot}} (\{r_i\}), \text{ starting from } \{r_i\} = \{r_i(t)\},
\]

(8)

where \( \{r_i\}_{\text{free}} \) are the positions of the free atoms, i.e. all but the atoms in the fixed bottom layers and the atoms of the indenter. Equations (5) and (7) can be restated in incremental form,

\[
\Delta E_{\text{tot}}(t) = \Delta E_{\text{kin}}(t) + \Delta E_{\text{pot}}(t),
\]

(9)

\[
\Delta E_{\text{pot}}(t) = \Delta E_{\text{pot,therm}} + \Delta E_{\text{pl}},
\]

(10)

where \( \Delta E_{\varsigma}(t) = E_{\varsigma}(t) - E_{\varsigma}(0) \).

Note that the sum of \( \Delta E_{\text{kin}} \) and \( \Delta E_{\text{pot,therm}} \) corresponds to the heat \( Q \) which is created by the friction phenomenon. Both \( E_{\text{kin}} \) and \( E_{\text{pot,therm}} \) are zero at equilibrium, therefore \( E_{\text{pl}} \) becomes

\[
E_{\text{pl}}(t) = \min_{\{r_i\}} E_{\text{pot}}(t) - E_{\text{pot}}(0).
\]

(11)

Note that the truth of (11) depends on the quenching’s finding the closest local minimum, i.e. that the quenching does not modify any plastic phenomena. We used a conjugate gradient (CG) method and a visual inspection of the dislocation patterns such as those shown in Figure 3 before and after quenching suggested that this condition is satisfied by CG.

2.3. Simulation volume

Table 1 lists all the simulations required to execute this parametric study. Note that due to the MS simulations at each snapshot, a large number of individual computations has been performed. Their management and organisation of the output data are challenges of their own. The largest simulations performed in this study are the ones for \( v = 10 \frac{m}{s} \) and \( h = 183.1 \text{ Å} \) with a total of 789,466 atoms for 3,000,001 time steps.

\(^2\)The atomic velocities \( \{r_i(t)\} \) are not considered in molecular statics and set to zero.
Fig. 3. Snapshots of dislocation patterns during the simulation at four different indenter positions \( l \). The indenter atoms are represented by blue spheres, red spheres represent dislocated atoms in the bulk and white spheres belong to chip and groove atoms. All other atoms are represented by transparent white dots. The substrate thickness is \( h = 91.5 \) Å, the indentation depth is \( \Delta y = 2 \) Å and the scratching speed is \( v = 10 \) m/s. For clarity, only the dislocated atoms belonging to the two mentioned loops are represented.
3. Simulation results

3.1. Work and plasticity

Figure 4(a) shows plots comparing the evolution of \( W_{sc} \) to the evolution of \( E_{pl} \) for three different substrate thicknesses at a scratching speed \( v = 10 \text{ m s}^{-1} \) for grazing contact \( \Delta y = 0 \). Deeper indentation depths yield similar evolutions with higher energies involved. We can see that both \( W_{sc} \) and \( E_{pl} \) rise essentially monotonically (save for high frequency fluctuations) with increasing scratch distance. Most importantly, we see a significant size effect; both \( W_{sc} \) and \( E_{pl} \) are significantly higher for the thicker substrate. Note that \( E_{pl} \) is a non-negligible proportion of the total scratching work contributing to roughly one tenth of the energy increase at this speed. The difference between \( W_{sc} \) and \( E_{pl} \) is the heat generated by the scratch. Figure 4(b) shows similar plots but for which \( h = 22.9 \text{ Å} \) has been kept constant and the scratching speed is varied. While there is a distinct rate dependence in \( W_{sc} \), especially at higher scratch speeds, \( E_{pl} \) appears to be insensitive to the rate. This implies that for range of speeds covered in this study, \( E_{pl} \) is a simulation box size dependent phenomenon, while \( W_{sc} \) is both size and rate dependent.

![Graphs showing work and plastic energy](image)

(a) \( v = 10 \text{ m s}^{-1} \) for different substrate thicknesses \( h \). Heavier lines stand for thicker substrates. Solid and dashed lines show the scratch work \( W_{sc} \) and the plastic energy \( E_{pl} \), respectively. Note the size dependence of both \( W_{sc} \) and \( E_{pl} \).

(b) \( h = 22.9 \text{ Å} \) for different scratch speeds. Solid and dashed lines show the scratch work \( W_{sc} \) and the plastic energy \( E_{pl} \), respectively. Note that \( W_{sc} \) features rate dependence while \( E_{pl} \) is somewhat insensitive to varying \( v \) in the speed range of this study.

![Graphs showing work and plastic energy](image)

Fig. 4. Grazing contact (zero indentation depth): comparison between scratching work \( W_{sc} \) (solid lines) and plastic energy \( E_{pl} \) (dashed lines). Note that \( W_{sc} \) and \( E_{pl} \) share the same abscissa. The values of \( E_{pl} \) are to be read on the right ordinate, those for \( W_{sc} \) on the left one.

Table 1. Individual simulations performed for the parametric study, where \( N_h, N_v, N_{\Delta y} \) are the number of substrate thicknesses, scratching speeds and indentation depths, respectively, \( N_t \) is the number of runs performed with a thermostat for reference (see footnote) and \( N_c, N_s \) are the total number of distinct scratch cases and of analysed points per case, respectively.

<table>
<thead>
<tr>
<th>Type</th>
<th>Number of simulations</th>
</tr>
</thead>
<tbody>
<tr>
<td>indentation MD</td>
<td>4 ( N_h )</td>
</tr>
<tr>
<td>indentation minimisation</td>
<td>4 ( N_h )</td>
</tr>
<tr>
<td>scratch MD</td>
<td>45 ( N_c = (N_h + N_v - 1) \times N_{\Delta y} + N_t )</td>
</tr>
<tr>
<td>scratch minimisation</td>
<td>13545 ( N_c \times N_s )</td>
</tr>
<tr>
<td>total</td>
<td><strong>13598</strong></td>
</tr>
</tbody>
</table>
One can discern a transient regime in the first 10 to 15 nm in which $W_{sc}$ grows like a parabola, followed by what appears to be a permanent regime in which $W_{sc}$ grows linearly. This effect can be explained by the developing plastic zone under the indenter: early dislocations can easily travel into the still perfect crystal (see Figure 3(b)) while later dislocations tend to pile-up against the fixed bottom layers of the crystal (see Figure 3(d)) and harden it, hence the increasing slope of $W_{sc}$ during the transient regime. Once the pile-up is reached, the plastic zone cannot keep growing and permanent regime is reached. The size effect we observe is a reflection of the inability of the plastic zone to develop fully.

A comparison between the number of dislocated atoms $N_{pl}$ and $E_{pl}$ is shown in Figure 5 for $h = 183.1$ Å and grazing contact. Note that the two plots share the same abscissa, but have different ordinates on either side of the graph.

One can observe that $E_{pl}$ is generally smoother than $N_{pl}$, which is not surprising since $E_{pl}$ computes a continuous result of a global measure, whereas the count of dislocated atoms is based on categorical data; either the centrosymmetry parameter $P$ is above threshold or not. While $E_{pl}$ increases continuously during the nucleation of any dislocation loop, the categorical nature of $N_{pl}$ can be clearly seen in the way it rises in discrete plateaus (superposed to some noise, i.e. upwards spikes). See the transitions at about 16 nm, 18 nm etc. in Figure 5 which represent the nucleation of distinct dislocation loops. The good fit between the two plots is evident and shows that $E_{pl}$ indeed is a satisfactory quantitative measure for the energy stored in plastic zones.

3.2. Friction coeﬃcient

The most common characterisation of friction [23] is through the friction coefficient $\mu$ defined as [8]

$$\mu \equiv \frac{dF}{dL},$$

(12)

where $F$ is the friction force in the scratching direction and $L$ is the normal force. The definition is based on the assumption that $F$ is a rate-independent affine function of $L$ of the form

$$F = F_a + \mu L,$$

(13)
where $F_a$ is an adhesion force which depends on the contact area and $\mu$ is a parameter describing the interaction between the indenter and substrate surfaces.

The friction coefficient is a macro-scale concept and requires some adaptation in order to be meaningful at the nano-scale; the forces $F(t)$ and $L(t)$ can be extracted from the simulations, but they fluctuate very strongly due to high frequency thermal vibrations and make a fit to (13) in order to estimate $\mu$ meaningless. An instantaneous friction coefficient $\mu(t)$ would defy the purpose of the concept. Conversely, getting rid of all fluctuations by averaging the forces over entire simulations

$$\bar{F} = \frac{1}{N} \sum_{i=N}^{N_i} F(t_i),$$

$$\bar{L} = \frac{1}{N} \sum_{i=N}^{N_i} L(t_i),$$

where $N$ is the total number of simulated time steps yields representative estimations of both $F$ and $L$ which can be fitted to (13). However, it makes it impossible to determine confidence intervals for $F - L$ data points. We propose a floating window averaging of the forces

$$\langle F \rangle_i = \frac{1}{N_w} \sum_{i=0}^{N_w} F(t_{i+j}),$$

$$\langle L \rangle_i = \frac{1}{N_w} \sum_{i=0}^{N_w} L(t_{i+j}),$$

$$i \in \{0, 1, 2, \ldots, N - N_w\},$$

where the window size $N_w$ is given by

$$N_w = \frac{a_s}{v \Delta t},$$

where $a_s$ is a lattice distance in the scratching direction and $\Delta t$ is the simulation time step. The window size corresponds to the time it takes the indenter to move over one periodic unit cell of length $a_s$ of the substrate crystal. This way, each window-averaged data point represents forces clean from fluctuations of higher frequencies than those which are crystallographically explicable.

Figure 6(a) shows $\langle F \rangle$ as a function of $\langle L \rangle$ for different scratch speeds $v$. The speeds 10 and 40 m/s are not shown in Figure 6(a) for clarity but do not differ substantially from the rest. Each plotted data point represents a window-averaged force measurement cluster for an entire scratch simulation and the error crosses show their standard deviations which are taken to be confidence intervals. One cluster has been computed for each scratch case in the parameter space $P_v$

$$P_v = \{v\} \times \{\Delta y\},$$

$$\{v\} = \{5, 10, 20, 40, 80\} \text{ m/s},$$

$$\{\Delta y\} = \{0, 1, 2, 5, 10\} \text{ Å}$$

for $h = 22.9 \text{ Å}$. The straight lines are obtained from least-squares linear approximations for clusters of equal scratch speed and their slope is the friction coefficient $\mu$, see Definition (12). One can see that all linear approximations intersect the confidence intervals of all clusters. We conclude that there is indeed an affine relationship between $\langle F \rangle$ and $\langle L \rangle$ and that the slight rate dependence visible in the plot of $\mu$ as a function of $v$ (see Figure 6(b)) is not statistically significant.

Figure 7(a) shows the result of a similar analysis in which the parameter space $P_h$

$$P_h = \{h\} \times \{\Delta y\},$$

$$\{h\} = \{22.9, 45.8, 91.5, 183.1\} \text{ Å},$$

$$\{\Delta y\} = \{0, 1, 2, 5, 10\} \text{ Å}$$

for $v = 10 \text{ m/s}$ is explored. Statistically, the affine relationship between $\langle F \rangle$ and $\langle L \rangle$ is still valid, with every linear approximation intersecting the confidence intervals of their corresponding clusters. It seems at a first glance at Figure 7(b) that the thick substrates (in this case, the ones for which $h \geq 45.8 \text{ Å}$) are large enough to overcome size effects, i.e., $\mu$ exhibits a size dependence only in the thin substrate case, where the
indentation depths are comparable to the thickness. The confidence intervals, however, have significantly grown with respect to the previous case, especially for the clusters with high averaged forces. It is therefore not obvious to conclude size-independence for large thicknesses based on the global friction forces.

Figure 8 compares the relative evolution of plastic energy $E_{pl}$ as a function of the thickness $h$ for two different indentation depths. For both indentation depths, the evolution of $E_{pl}$ appears stochastic and no trends can be seen, but in both cases, $E_{pl}$ varies considerably and non-monotonically. While the friction coefficient $\mu$ (and therefore the involved scratching work $W_{sc}$) stabilises for large $h$ and suggests that sufficiently large simulation box sizes have been reached, the plastic energy $E_{pl}$ reveals that the underlying mechanisms and plastic effects are still size-sensitive.

Note that a) the scratch speeds $v$ in the simulations are quite high and that the heat production $Q$ during scratching decreases with decreasing $v$, and b) that our results suggest that $E_{pl}$ is insensitive to $v$. This entails that $E_{pl}$ becomes a more important and determining quantity at lower $v$.

It is safe to conclude that the parametric study has not covered simulation box sizes large enough for friction mechanisms to develop freely. Of course, it is possible to increase the size further and to perform larger computations, but it is important to note that the presented scratch simulations treat the case of a very small indenter at shallow indentation depths. As a comparison, the typical tip radius of an atomic force microscope (AFM) is 100 nm [24] – over an order of magnitude larger than the indenter in the present case. An AFM-size simulation would easily fall back in the fully size-dependent case where the indentation depths are comparable to the substrate thickness or be computationally extremely costly. Understanding the mechanisms in play with nano-scale friction will require the investigation of substantially larger systems very likely to be out of reach of pure MD simulations. Therefore, the investigation of friction at the atomic scale calls for a rigorous multi-scale treatment.

Several types of multi-scale methods have been developed for similar cases;

- Sequential multi-scale studies like Chang et al. [16] start problems in MD and switch to DD once dislocation network nucleation has happened. Such studies yield good results for indentation problems. However, they are not suited for frictional (i.e. sliding, scratching) contact because they raise problems when dealing with the continuing nucleation of new dislocation networks for the entire simulated
• Concurrent multi-scale methods with predefined atomistics-continuum interfaces such as the bridging domain method [25, 26, 27] solve the problem only partly because the interfaces are insurmountable obstacles to dislocation motion and trap plasticity within the atomistic domain.

• The quasicontinuum (QC) method circumvents this problem by expanding the atomistic domain as dislocations approach the interface. 3D QC calculations, however, become prohibitively costly to continue when larger and larger dislocation networks develop and QC comes closer and closer to single-scale MD.

• Finally, the coupled atomistics and discrete dislocations (CADD) method [28, 29] couples MD concurrently to DD and finite elements with a predefined atomistics-continuum interface but provides a mechanism for dislocations to transcend from an atomistic description in MD into a discrete description in DD and vice versa. It is to our knowledge the only atomistics-continuum multi-scale method allowing plasticity at dislocation resolution to nucleate atomistically and to expand beyond the atomistic domain. However, CADD has only been implemented in 2D.

We conclude that understanding the atomistic origins of friction in crystalline metals requires a 3D CADD implementation or a 3D CADD-like method which allows plasticity to nucleate in MD and expand over larger domains than what is treatable in MD on present-day computing hardware.

4. Conclusion

We have performed a large parametric molecular dynamics study of single-asperity single-crystal aluminium nano-scratch simulations in which we varied the substrate thickness, the indentation depth and the scratch speed over more than an order of magnitude each. The indenter size is about an order of magnitude smaller than a typical atomic force microscopy tip. Using a novel combined molecular dynamics and molecular statics approach for analysing nano-scale friction, we have been able to define a clean separation of the
friction work $W_{sc}$ into heat generation $Q$ and creation of permanently stored plastic deformation energy $E_{pl}$. By comparing the evolution of the number of dislocated atoms $N_{pl}(t)$ to the evolution of the plastic energy $E_{pl}(t)$ we obtained strong indications that our method to compute $E_{pl}$ is indeed able to accurately quantify the energy associated with plastic zones independently of how they have been created.

The generated heat $Q$ has been found to be distinctly scratch speed dependent, while – for the range of speeds considered – $E_{pl}$ shows only weak speed sensitivity. Both $Q$ and $E_{pl}$ have been shown to feature a distinct sensitivity to the simulation box size.

We have presented a regression-based method to compute friction coefficients $\mu$ and defined a window-average for measurements extracted from the simulations. This window-average provided us with a statistically motivated criterion to determine that – for the range of scratch speeds considered – there is no significant speed sensitivity of $\mu$. The same criterion has shown, however, that $\mu$ is simulation box size dependent in a complex manner: the average value of $\mu$ seems to stabilise for substrates thicker than about five times the indentation depth, but its confidence interval grows large. The associated $E_{pl}$ does not stabilise, indicating that the plastic zones are not resolved, even for the relatively small scratch case covered in this parametric study.

In order to understand the atomic origin of friction at engineering size scales or at least at the scale of atomic force microscopy, a rigorous multi-scale treatment of the problem becomes necessary. A 3D CADD implementation is currently being developed by the author of the original CADD method and the authors of this document.

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