Simulation of copper nanoindentation using LAMMPS - atomic to micro-scale

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Abstract

This study explores the mechanical properties of copper at the nanoscale through molecular dynamics simulations of nanoindentation using LAMMPS. Focusing on atomic-scale interactions, the research investigates the relationship between indentation depth and pressure, considering factors like indenter shape and application rate. The simulations reveal complex material behaviors diverging from classical Hertzian contact theory, highlighting the intricacies of nano-scale mechanical responses. The study contributes significantly to the understanding of copper's mechanical properties and the application of computational simulation in material science.

Keywords: *Nanoindentation*, *Molecular Dynamics Simulation*, *Copper Mechanical Properties*, *LAMMPS*, *Nanoscale Material Behavior*, *Atomic-Scale Interactions*, *Computational Material Science*

1 Introduction

Copper is a widely used metal with various industrial applications due to its excellent electrical and thermal conductivity, making it essential for electronics and energy applications. However, its mechanical properties at the nanoscale can differ significantly from its bulk properties due to size effects (Meyers, Mishra, & Benson, 2006a, 2006b).

The mechanical properties of materials at the nanoscale can be measured by nanoindentation. Nanoin-

dentation, that uses a nano-indenter with a known tip shape penetrates the specimen at a specific location by applying user defined load, has been considered one of the most powerful methods to assess the mechanical properties at micro/nano scale (Voisin, Krywopusk, Mompiou, & Weihs, 2017). With this, material parameters like hardness, elastic constant, yield stress, and residual stresses can easily be obtained by using the results of nanoindentation (Oliver, Bradby, Williams, Swain, & Munroe, 2009; Zeilinger et al., 2016). This technique is crucial for understanding the mechanical behavior of materials in various applications, such as microelectronics, coatings, biomaterials, and more. However, there are many difficulties in conducting experiments at the nanoscale.

Computational simulations for nanoindentation have been widely used to resolve the difficulties that appeared during experimentation at the nanoscale. Molecular dynamics (MD) is one of the most powerful and efficient tools to investigate the mechanical properties of materials like monocrystalline and polycrystalline (grain size ranges from 1 to 100 nm) materials (Kim et al., 2018; Li et al., 2016; Roy, Sharma, Ranjan, & Balasubramaniam, 2021).

Typically, nanoscale copper has been considered as a well-suited material for investigating the mechanical properties using MD (Chang, 2003; Sharma, Joshi, Datta, & Balasubramaniam, 2020). Huo et al. (Huo, Liang, & Cheng, 2007) fabricated monocrystalline copper thin film using the vapour deposition method and performed a nanoindentation test to assess hardness and elastic modulus. They showed that hardness depends on the size of the indenter and the depth of indentation. Chao-Chun et al. (Huang, Chiang, & Fang, 2015) studied the nanoindentation based mechanical performance of monocrystalline and polycrystalline copper using the MD approach. Schiotz et al. (Schiøtz & Jacobsen, 2003) studied the mechanical properties of polycrystalline copper under the nanoindentation process. They observed maximum flow stress and strength in polycrystalline copper with a 10–15 nm grain size range (Armstrong, 1987; Hall, 1951).

In this study, we employed LAMMPS to perform molecular dynamics simulations on atomic-scale copper nanoindentation. We established a correlation between the depth of indentation and the pressure at the designated material position. Additionally, we explored the influences of the indenter's shape and the rate of application on the connection between indentation depth and pressure.

2 Methodology

We approached the experiment with a pragmatic mindset, meaning we believed that knowledge could be gained through unbiased observation and measurement. To simulate the process of the nanoindentation between the copper surface and the indenter in a 3D classic molecular dynamics manner, we are currently using the open-source code, LAMMPS, to simulate the behaviour and effect of copper, study the underlying mechanism of the copper material, and achieve the visualization in OVITO, an open visualization tool on an atomic scale. The configuration of the copper coating is simulated by an embedded atom method (EAM) potential in which the potential is given by the following equation

$$E_i = F_\alpha \left(\sum_{i \neq j} \rho_\beta (r_{ij}) \right) + \frac{1}{2} \sum_{i \neq j} \phi_{\alpha\beta} (r_{ij}) \tag{1}$$

where r_{ij} is the distance between atoms i and j, $\phi_{\alpha\beta}$ is a pair-wise potential function, ρ_β is the contribution to the electron charge density from atom j of type β at the location of atom i, and F_α is an embedding function that represents the energy required to place atom i of type α into the electron cloud (Ahadi, Hansson, & Melin, 2016). The ability of EAM potentials to reproduce phase diagrams is very important for many applications of atomistic simulation (Williams, Mishin, & Hamilton, 2006). For our research, we have chosen to utilize the Cu_u3.eam potential file provided by LAMMPS as a foundational tool for determining copper's potential. To

ensure that the copper atom does not come into contact with the indenter's fictive surface, we must use the "indent" built-in function in LAMMPS. Additionally, one surface is free while the other is fixed for indentation. This means that the bottom of the copper must be restricted as a constraint. When designing the parameters, we have set the temperature of the simulation as a constant T=300K, with a time step of 0.002ps. You can find more detailed information about the parameters in Table 1 below.

Table 1: Parameters

Relaxation steps	0.0002ps	Temperature,1	300k
Time step	0.2ps	Lattice parameter, a_0	3.615 Å
Maximum indentation depth	$54a_0$	Indentation steps	80000
Indenter velocity	10Am/ps and 20Am/s	Number of atoms V	24800

In our research, the objective was to establish a reliable depth-force relationship that adheres closely to the Hertzian contact theory. To achieve this, we employed the LAMMPS molecular dynamics simulator and implemented a series of calibration procedures to refine our simulations and ensure reliable results. To accurately represent real-world conditions, we focused on adjusting the simulation temperature. Recognizing the influence of temperature on atomic behavior and depth-force characteristics, precise temperature calibration was essential. We fine-tuned the simulation temperature to ensure that the atomic interactions were representative of real-world conditions. In addition to temperature, we considered the total number of atoms in the simulation. Striking the right balance, we aimed to capture the nuances of the depth-force interaction without oversimplifying or overcomplicating the system, while ensuring computational feasibility. Meticulous attention was given to setting appropriate boundary conditions. By employing periodic boundary conditions across all dimensions, we eliminated edge effects that could introduce bias and distort the depth-force relationship. Following the calibration procedures, we generated a depth-force curve from the simulation results. This curve was then compared against the theoretical Hertzian contact formula, serving as a validation step to assess the reliability of our simulation settings. To mitigate potential errors, we gradually adjusted the boundary conditions, atomic formulas, and temperature, while increasing the number of atoms. Through this iterative process, we aimed to obtain simulation conditions that closely aligned with the Hertzian contact formula, validating the accuracy of our fitting approach.

To verify the soundness of this simulation and to show that it is a self-consistent system, we use the following equation:

$$\frac{1}{E_{tot}} = \frac{1}{E_{tip}} \frac{1}{E_{surf}} \tag{2}$$

$$a = \sqrt[3]{\frac{RF}{E_{tot}}} \tag{3}$$

$$\delta = \frac{a^2}{R} = \left(\frac{F^2}{RE_{tot}^2} \right)^{1/3} \tag{4}$$

δ:contact compression
F:contact force
a:actual contact radius
R:radius of indenter
E_{tot}:total Young's modulus

Note that we don't know the constants such as E_{tot} in these two expressions, but we can observe the power series relationship between δ and F in the data group. Through these methodical adjustments, we were able to

mitigate potential errors and ensure a robust simulation framework. This rigorous approach bolstered our confidence in the simulation's ability to replicate real-world phenomena and validated the integrity of our findings. Our findings contribute to the advancement of depth-force characterization in materials science research.

3 Results

Contact Behavior Analysis: From the molecular dynamics simulation, the relation between depth (d) and force (F) during the atomic scale indentation process was observed. The data suggest a non-linear relationship that does not immediately align with the Hertzian contact theory. The results range from an indentation depth of 0 to 15 units (exact unit not specified) with the force varying significantly, starting at 0 and reaching a maximum of around 166 units.

System Initialization: The LAMMPS script delineates a system with metal units in a three-dimensional periodic boundary condition. Two distinct atomic regions were defined: a "base" and a "tip". The "base" region uses type 1 atoms formed in an FCC lattice, while the "tip" is defined as a sphere using type 2 atoms, also in an FCC lattice. An equilibrium run of 70,000 timesteps is executed to stabilize the system, with the "base" region undergoing NVT ensemble conditions to maintain a temperature of 300K.

Ovito Visualization Characteristics: Visualization of the simulations through the Ovito software unveiled intricate atomic-scale dynamics during the indentation process. As the depth of indentation increased, dislocation events became prominent. Atoms involved in these dislocations emanated in a wave-like pattern outward from the site of indentation. This wave-like expansion of dislocated atoms vividly depicted the strain propagation through the material. Furthermore, on the horizontal plane, the length or stretch of dislocated atoms grew, suggesting a wider zone of influence or perturbation as the indenter drove deeper into the material.

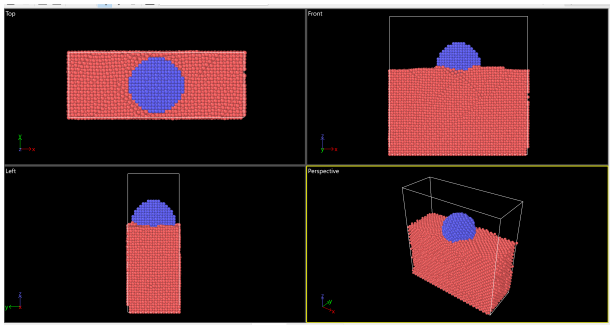

Figure 1: The visualigasim OVITO

Force-Depth Curves & Hertzian Contact Theory Fitting: The force-depth curves obtained from the simulations exhibited distinct characteristics. Initially, when examining the first 50 data points, the curve showed a trend where it initially approached a horizontal asymptote. However, as we further explored beyond the 50th data point, the curve displayed an unbounded increasing trend. When we attempted to fit our force-depth curve with the Hertzian contact theory, the fit was not perfect, suggesting that the actual interatomic interactions and responses could be more complex.

Parameter Variations Impact: Changing the timestep and velocity parameters in the LAMMPS simulations produced noticeable variations in the force-depth curves. Specifically, increasing the indenter's velocity led to a higher force response in the material. Additionally, the curves exhibited different tendencies based on these parameter adjustments.

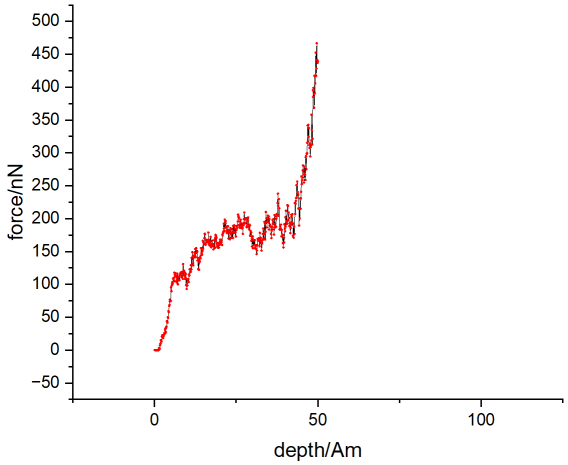

Figure 2: N_0=500,v_0=10Am/ps

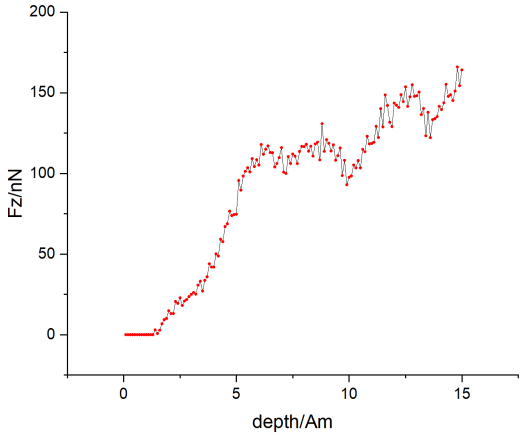

Figure 3: N_0=100,v_0=10Am/ps

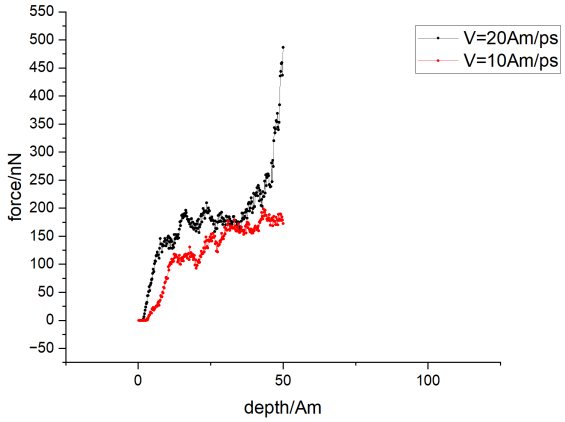

Figure 4: N_0=500,v_0=20Am/psN_0=500,v_0=10Am/ps

4 Discussion

Nanoindentation stands as a valuable technique for delving into the intricate mechanics of materials at the nanoscale. The insights from previous discussion offer a thought-provoking perspective on the complexities and consequences of nanoindentation outcomes.

Through analyzing the disparities between simulated force-depth curves and predictions based on Hertzian contact theory, we can gain valuable insights into the intricate nature of material behavior at small scales. While Hertzian contact theory provides a fundamental understanding, it oversimplifies the complex interatomic interactions that occur during nanoindentation experiments. Therefore, it is crucial to consider factors such as atomic adhesion, surface roughness, and atomic-scale heterogeneities which can significantly deviate real-world behaviors from classical projections.

A notable aspect comes to light when considering the influence of indentation velocity on force-depth curves, highlighting the significance of accounting for the time-dependent aspects of material response. The noted increase in resistance and amplified forces at higher indentation velocities can be attributed to the constrained time frame for atomic reorganization and stress redistribution. Rapid indentation impedes material relaxation, resulting in heightened resistive forces.

Furthermore, investigating the implications of adjusting simulation parameters, such as the timestep, can provide deeper insights into the level of detail captured during molecular dynamics simulations. Employing a smaller timestep yields a more intricate representation of atomic transitions but requires greater computational resources. Conversely, using a larger timestep may compromise precision in favor of computational efficiency.

The sensitivity of these parameters emphasizes the pivotal role played by selected factors, such as velocity and timestep, in shaping nanoindentation simulations. These variations directly influence the mechanical responses exhibited by materials, thereby highlighting the crucial importance of meticulous parameter selection to ensure accurate depiction and prediction of material behaviors observed in real-world scenarios.

It is imperative to approach the interpretation of nanoindentation data with utmost caution. While nanoindentation offers invaluable investigative potential, it is essential to acknowledge the inherent limitations of both simulations and experimental outcomes in capturing the intricate complexity of nanoscale material behavior. Accounting for factors such as surface effects, adhesion phenomena, and the dynamic evolution of processes

becomes indispensable when drawing meaningful and accurate conclusions about material properties.

As nanoindentation techniques continue to advance, there is potential for the incorporation of advanced models that capture atomic-scale interactions and complexities, thereby bridging the gap between simulations and theoretical predictions. Promising directions include integrating molecular dynamics with continuum mechanics or incorporating quantum mechanical effects, which could enhance prediction accuracy and deepen our understanding of mechanical behavior at the nanoscale.

5 Conclusion and Outlook

In our research, we employed the LAMMPS molecular dynamics simulator to model and examine the system. Recognizing the inherent deviations and discrepancies in computational simulations, a series of calibration procedures were instituted. Initially, we fine-tuned the simulation temperature to ensure that the atomic interactions were representative of real-world conditions. Subsequently, we adjusted the total number of atoms involved in the simulation. Furthermore, boundary conditions were meticulously set. We adopted periodic boundary conditions across the three dimensions to eliminate edge effects that might skew the depth-force relationship. Following these calibrations, we generated a depth-force curve from the simulation results. The curve was then cross-examined against the Hertzian contact formula. A close match between the simulated curve and the theoretical Hertzian curve would indicate the reliability and validity of our simulation settings. From the correlation we established, we can know that the depth of indentation and the pressure conform a kind of algebraic relationship, which can help us learn more about copper, and software such as LAMMPS play a key role in the research. Here are some key conclusions:

1. Magnitude of force largely determines the accuracy of the simulation; when the force is not strong enough, the curve shows a trend of approaching a horizontal asymptote, with the rise of the force, the depth of the material will show a near-unbounded escalation. This may be due to Onset of Plastic Deformation and Breaking of Stronger Bonds.

2. When the indenter has not yet come into contact with the surface, because of electromagnetic force, atoms may be pulled, so the function image may not pass through the coordinate origin.

3. There are many factors such as temperature and forces that can influence the function curve, but these curves may not be fitted to elementary functions, so the dataset we get using software may help the research of others.

As for the outlook on simulations of nanoindentation; nanoindentation of conventional metallic materials is a relatively mature technology, so often we do not have to use software, and copper is a kind of conventional metallic material, but in other areas, such as nanoindentation of polymer composite materials and their constituents, it continues to be a very active and difficult research area. Research on hybrid atomistic simulations of nanoindentation has been focused mainly on metallic and ceramic materials, but it would seem that such simulations would also be beneficial in studies of polymer composites, especially the interphase region. Moreover, there are many reports in the literature on the use of micro- or nanoindentation push testing to measure a material such as the IFSS of traditional fiber reinforced polymers. In this area nanoindentation is hard to implement in experiments, using nanoindentation to measure the IFSS of nanotube reinforced polymers seems to be an undeveloped research topic. Therefore, in the future, computational simulations for nanoindentation will bring us further development.

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Author Contributions

Conceptualization: J.Z., Y.H., D.L. and Z.L.; methodology, validation, formal analysis: M.T., J.H., D.L. and Z.L.; investigation, resources, data curation: M.T., J.H. and J.Z.; writing-original draft preparation, visualization: M.T. and D.L.; writing-reviewing and editing, visualization and supervision: Y.H and D.L.; Project administration: J.Z., M.T. and D.L. All authors have read and agreed to the published version of the manuscript.

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Informed Consent Statement

Informed consent was obtained from all subjects involved in the study.

Data Availability

Please contact the corresponding author for all reasonable requests for access to the data.

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Conflicts of Interest

The authors declare no conflict of interest.

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