Research Article

Molecular Dynamics Simulation of Nanoindentation of Cu/Au Thin Films at Different Temperatures

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Two methods, deposition method and ideal modeling based on lattice constant, are used to prepare three modulation periods' (1.8 nm Cu/3.6 nm Au, 2.7 nm Cu/2.7 nm Au, and 3.6 nm Cu/1.8 nm Au) thin films for nanoindentation at different temperatures. The results show that the temperature will weaken the hardness of thin films. The deposition method and the formation of coherent interface will result in a lot of defects in thin films. These defects can reduce the residual stress in the thin films which is caused by the external force. The proposed system will provide potential benefits in designing the microstructures for thin films.

1. Introduction

Coatings play important roles in the modern manufacturing and processing industry [1]. The tools and engines obtain certain characteristics (e.g., super-hardness, excellent antiwear capability, and high heat-resistance) by coating specific materials on the surface. Recently, nanomultilayer coatings (NMLCs) [2] have drawn significant attentions for their multifunction applications in comparison to monolayer coatings, as the NMLCs are promising candidatures to use in the field of aerospace, manufacture, automobile, and so on.

Several novel experimental methods (e.g., nanoindentation, nanoscratch) were proposed to examine the properties of NMLCs. Li et al. [3] found two different deformation mechanisms, that is, elastic buckling-assisted grain boundary sliding and dislocation plasticity-dominated shearing, of Au/Cu multilayer via nanoindentation tests. However, the micromechanisms of NMLCs are difficult to reveal by these experiments. Therefore, analytical models [4] and numerical methods [5–7] were used to cover the shortage of experimental measurement.

Molecular dynamics (MD) simulations [8] are developed based on the classical Newton and statistical mechanics, and have become a powerful tool to investigate a serial of complex systems [9–12] or conditions [13–17], which can be widely

applicable in the area of nanomaterials. Especially, MD can be used to examine the materials in extreme conditions. Landman et al. [18] used a nickel tip to perform the indentation of a gold surface by MD. The gold surface experienced onset of plastic deformation. Zientarski and Chocyk [19] examined the structure and stress in Cu/Au and Fe/Au thin films. The compressive stress was observed in Cu/Au thin films while tensile stress was found in Fe/Au thin films.

Many interesting mechanisms of NMLCs had been revealed based on MD [20, 21]. However, the studies of coatings are still intensive because of the importance of their application. Meanwhile, there are certain thermal conditions (e.g., high temperature) that exist during the real working process of thin films. Thus, in the present paper, MD is employed to perform the nanoindentation of Cu/Au thin films at different temperatures, to investigate the detail molecular mechanisms regarding the preparation methods.

2. Model and Computational Method

2.1. Preparation of Cu/Au Thin Films. The preparation of Cu/Au thin films is based on the deposit system (PVD method) and is shown in Figure 1. The simulation box is $7.2 \times 7.2 \times 30 \text{ nm}^3$ in $X \times Y \times Z$ dimensions. The Au atoms are randomly generated in the region of 25–27.5 nm in Z -axis.

FIGURE 1: Simulation model system of deposition.

Then, Au atoms deposit on Cu-substrate with the incident kinetic energy of 0.15 eV.

The substrate in Figure 1 consists of 16,000 Cu atoms which is $20 \times 20 \times 10$ unit cells. This system is adapted from solid-liquid interface model [20, 22, 23]. The atoms at bottom two layers of substrate are fixed. Meanwhile, the other Cu atoms of substrate equilibrate in NVT ensemble at 300 K for 1 ns. Next, the atoms at top six layers of substrate are simulated in NVE ensemble for 1 ns, while the other atoms' interplaying remains unchanged. After that, Au atoms incident to the substrate in 1 ns with the deposition rate of 7.5×10^{24} atom/s·cm². The Au particles are simulated in NVE ensemble. While the Cu atoms in three circumstances, that is, NVE ensemble, NVT ensemble and fixed condition, interact with each other, as the Figure 1 presented.

Three modulation periods' thin films are obtained based on the model of deposition, which are 1.8 nm Cu/3.6 nm Au, 2.7 nm Cu/2.7 nm Au, and 3.6 nm Cu/1.8 nm Au. Also, the same modulation periods are used to generate ideal thin films without defect, based on the lattice constant of Cu and Au. The thin films studied in this work are denoted as Deposit 1.8, Deposit 2.7, Deposit 3.6, Ideal 1.8, Ideal 2.7, and Ideal 3.6. For example, Deposit 1.8 represents the 1.8 nm Cu/3.6 nm Au thin film prepared by the deposition model. The microstructures and thermophysical properties of these thin films had been discussed in our previous paper [20, 24].

2.2. Nanoindentation of Thin Films. The six obtained thin films are annealed (heated to 1,000 K and equilibrated for a short period, then cooled to 300 K) firstly. Then some atoms of the samples were removed to flatten the sample surface, for that the surface of the deposited sample is not flat. Next, the thin films interacted with an ideal spherical indenter with radius of 2 nm at a constant velocity of 50 m/s in Z-axis to obtain the load-depth (loading and unloading) curves. The parameter of radius and the impact velocity had been studied in another paper [6]. The load-depth is 1.6~2.2 nm for each thin film. Besides, the nanoindentation of these thin films is tested at temperatures of 0, 300, 700, and 1000 K, respectively. It should be noted that residual stress remains in thin film during the preparation. The goal of this paper is to reveal the atomic mechanisms in thin films regarding the preparation

methods. We assumed there is no residual stress in the film before indentation.

2.3. Forcefield and Computational Parameters. MD simulations are performed by using LAMMPS (large-scale atomic/molecular massively parallel simulator) [25]. The EAM forcefield [26] is chosen as the interaction potential. Periodic boundary conditions are applied in X and Y coordinates. Two ideal walls are created perpendicular to Z-axis at bottom and up surfaces of the simulation box as the boundary. The time-step is set as 1 femtosecond (fs) in the simulations. This time-step is tested for energy conservation. The Langevin method is employed to conserve the temperature [24].

3. Results and Discussion

The different microstructures will result in different properties of materials. The discussion of this paper will focus on the mechanical properties and deformation mechanisms of these thin films at different temperatures, since the microstructures of these thin films had been analyzed previously [20, 24].

3.1. Indentation Force-Depth Curves. The loading and unloading process of indentation can be illustrated by the force-depth (P - h) curves, which is the relationship between the force calculated from the indenter and depth of the indenter. The P - h curves of the investigated thin films are presented in Figure 2. Typically, in Figure 2(a), OA curve represents the loading process whose indenter moves into the thin films. And AB curve represents the unloading process whose indenter moves out of the thin films. The changes in these curves are as a result of the deformation in thin films.

Usually, P - h curves at 0 K are used to demonstrate the mechanism of indentation tested by MD. As shown in Figure 2, the force at 0 K is larger than that of other higher temperatures, that is, 300 K, 700 K, and 1000 K, even if the indenter reaches deeper depth. There is little difference of P - h curves among 300 K, 700 K, and 1000 K. However, there is a trend among the forces at different higher temperatures, which is $P_{300\text{ K}} > P_{700\text{ K}} > P_{1000\text{ K}}$. In other words, all of the P - h curves agree with the previous reports [27, 28] that the hardness decreases with the temperature increasing.

FIGURE 2: Indentation P - h curves for the deposit and ideal system.

FIGURE 3: Continued.

FIGURE 3: Continued.

(e) Deposit 3.6

(f) Ideal 3.6

FIGURE 3: The lattice evolution of thin films during nanoindentation at 0 K.

The atoms vibrate harder as the temperature increase. The atoms are easier moving by the external force at higher temperature. Thus, the force on the indenter is smaller at higher temperature.

3.2. Deformation Mechanism. The deformation of thin films during the nanoindentation can be visualized and illustrated by the lattice evolution. The face centered cubic (FCC) structure is the lattice structure for the pure Cu/Au metal. The indentation will move the atoms and form the defects in thin films. By visualization, this makes the lattice structure transform from FCC to other structures (such as amorphous, HCP, and BCC). Therefore, the other structures (except the FCC) are analyzed to study the deformation. And it should be noted that the lattice structure is complex and hard to distinguish by naked eye when the atoms vibrate at high temperature. Thus, the lattice evolution (without FCC) of thin films during nanoindentation at 0 K is presented as Figure 3. The lattice structure is visualized and analyzed based on the open software OVITO [29].

As discussed in our previous paper [20, 24], there are lots of defects in the thin films which prepared by deposition method. As shown in Figures 3(a), 3(c), and 3(e), there are lots of visualized lattice structures in the Deposit 1.8, Deposit 2.7, and Deposit 3.6 systems. And there are defects located at the coherent interface of Ideal 1.8, Ideal 2.7, and Ideal 3.6 systems. There are stacking faults in Deposit 1.8 system.

During the loading process, the indenter contacts with the thin film and causes the dislocation and slipping in the thin film. It should be noted that there are lots of defects in the thin film, and the dislocation and slipping is found under the stacking faults. The indenter did not pass through the Cu/Au coherent interface; however, the defects caused by the indenter transferred to the coherent interface. In the unloading process of Deposit 1.8 system, the defects reduced as the indenter removed from the thin film. However, there is residual stress in the thin film after the indenter reached its initial position. The residual stress will hold the atoms and hinder the thin film recover to its original configuration. The deformation process is clearer in the ideal systems.

In the Ideal 1.8 system, the indenter caused large defects in the thin film. The dislocation and slipping is more pronounced than that of the Deposit 1.8 system. And stronger residual stress remains in the thin films. It can be concluded that the defects caused by the deposition method could reduce the residual stress in a certain degree. The similar phenomena are also found in Deposit 2.7, Deposit 3.6, Ideal 2.7, and Ideal 3.6 systems. Interestingly, in the Deposit 3.6 and Ideal 3.6 systems, the indenter passed through the coherent interface and caused the defects in the Cu region. While these defects were eliminated after the indenter was removed from the thin films, as discussed earlier, there are defects in the coherent interface, which could also reduce or even eliminate the residual stress of the region under the coherent interface.

4. Conclusions

The Cu/Au thin films, which are obtained via deposition method and modeled based on lattice constant, with three

modulation periods (1.8 nm Cu/3.6 nm Au, 2.7 nm Cu/2.7 nm Au, and 3.6 nm Cu/1.8 nm Au) are employed to perform the nanoindentation test at different temperatures. The results could draw the following conclusions.

The hardness of the thin films is weaker as the temperature increase. The lattice evolution shows that thin films that obtained based on deposition method own more defects. And there are defects in the coherent interface of thin films. These defects could reduce the residual stress in the thin films after nanoindentation.

Competing Interests

The authors declare that they have no competing interests.

Acknowledgments

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