Computational Materials Science 81 (2014) 226–232

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

The influence of residual stress on incipient plasticity in single-crystal copper thin film under nanoindentation

Kun Sun [a], Weitao Shen [a], Lifeng Ma [b,*]

[a] State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, China
[b] S&V Lab, Department of Engineering Mechanics, Xi'an Jiaotong University, Xi'an 710049, China

ARTICLE INFO

Article history:
Received 1 June 2013
Accepted 12 August 2013
Available online 12 September 2013

Keywords:
Nanoindentation
Molecular dynamics
Dislocation
Copper

ABSTRACT

The incipient plastic deformation of single-crystal copper thin film with in-plane residual stress under Hertzian nanoindentation is studied using molecular dynamics simulation. The result reveals that the residual stresses significantly influence the surface strength and dislocation nucleation behavior of material: (i) the indentation hardness decreases with tensile residual stress, while it increases with moderate compressive residual stress, but it may drop down under higher compressive residual stress; (ii) the dislocation nucleation depth and its slip direction also vary with different residual stress state. This demonstrates that residual stress may influence incipient plasticity not only on the threshold value but also on the initial dislocation nucleation behavior under indentation.

1. Introduction

Residual stress can influence the material surface hardness and the incipient plasticity under indentation. In literatures, great efforts have been paid in this research [1–7]. In early 1930s, Kokubo [1] found that the applied in-plane tensile and compressive uniaxial stress has some very small influence on the Vickers hardness value. Similar results are also found by Sines and Carlson [2]. However, the observed effect of stress on the hardness is so small that no decisive conclusions could be drawn from these investigations. In 1996, Tsui et al. [3] studied the influence of the in-plane stress on the load–penetration depth curve and the contact area of indentation. They found that the hardness measured by sharp indenter increases with compressive stress, while it decreases with tensile stress. However, subsequent finite element simulation revealed that the change of hardness is not real, but rather an artifact caused by the pile-up of the surface, which cannot be accounted for in the nanoindentation data analysis according to the research result of Bolshakov et al. [4].

It seems that the existence of residual stress has a slight effect on the hardness and incipient plasticity measured by nanoindentation, while it has no effect on the measured elastic modulus up to now [5]. However, the problem is this: the indenters employed in these studies are sharp indenters (Vickers or conic shape). Once the sharp tip of indenters is pressing into the substrate surface, the plastic deformation of the substrate occurs due to the *singular* contact stress behavior. Then the dominant singular contact stress will largely diminish the influence of *regular* residual stress. Thus, sharp indenters possibly could not properly probe the residual stress influence on the incipient plasticity, the hardness, and the elastic modulus during indentation.

In comparison with sharp indenters, blunt indenters such as Hertzian indenter would produce regular contact stress, so the residual stress influence can be feasible to measure. Using spherical indentation, Taljat and Pharr [6] showed that residual stress has a clear effect on indentation load–penetration depth curve. By using molecular dynamics simulations, Schall and Brenner [7] demonstrated the importance of accuracy in the calculation or measurement of the projected contact area on the interpretation of nanoindentation results. Their conclusions are largely consistent with the findings of Tsui et al. [3].

In view of above different studies, it seems that the influence of residual stress on contact in literature is still not consistent and reconciled, and some advantages of blunt contacts are not fully explored. So it appears that further investigation of the influence of residual stress on the incipient plasticity, the local hardness, and the local elastic modulus under indentation, remains in order.

In this work, employing the Hertzian indenter, we conduct a series of molecular dynamics simulation of nanoindentation on single-crystal copper thin film with pre-existing equiaxial strain, to investigate the effect of residual stress on the incipient plasticity, the local hardness, and the local elastic modulus.

2. Simulation model and methodology

We simulate the nanoindentation process of single-crystal copper thin film along $\langle 1\,1\,1 \rangle$ growth orientation with a virtual sphere

* Corresponding author. Tel./fax: +86 29 8266 3861.
E-mail address: malf@mail.xjtu.edu.cn (L. Ma).

0927-0256/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.commatsci.2013.08.026

indenter of radius $R = 6$ nm using the public-domain computer software LAMMPS [8]. Fig. 1 is a schematic of a 2-D section of the nanoindentation configuration at $y = 0$. The x, y and z directions of simulation model are parallel to $[1\,1\,\bar{2}][\bar{1}\,1\,0][1\,1\,1]$ crystallographic directions, respectively. The models with and without pre-existing stress are of the same size $17.6 \times 13.2 \times 15.6$ nm, consisting of about 278,720 atoms, at initial configuration. An EAM potential, contributed by Mishin et al. [9], is adopted to model the Cu–Cu atoms interaction because it is calibrated according to the ab initio values of stacking fault and twin-formation energies.

The initial model of stress-free is relaxed using an energy minimization with a conjugate gradient method, followed by a zero-stress relaxation in the isothermal–isobaric ensemble (constant number of particles, pressure, and temperature, NPT) using a Nosé–Hoover thermostat at 0.1 K for 20 ps (20,000 steps). Then, pre-existing stress models are constructed by equiaxial compressing and/or tensioning the initial model in x and y directions at a constant strain rate $5 \times 10^5\ \mathrm{s}^{-1}$. After the desired strain level is reached, isothermal–isostrain MD simulation is performed in order to equilibrate the thin-film structure.

To study the effect of residual stress on the incipient plasticity, the indentation hardness, and the local elastic modulus of elastic substrate, the pre-strain values are adopted from -2% to 2% according to MD simulation result by Kolluri et al. [10]. In the case, the negative value means compressive strain while the positive means tensile strain. The corresponding pre-stress is displayed in Table 1, and normalized by the ideal shear strength ($G_{ideal} = 4.56$ GPa) for the Cu (1 1 1) single crystal in indentation experiment [11].

In the course of nanoindentation MD simulation, the substrate includes two kinds of atoms: boundary atoms and Newtonian atoms. Three layers of atoms at the bottom of the substrate (lower z plane) are boundary atoms, and the others are Newtonian atoms. The boundary atoms are constrained to $F_{normal} = 0$. The motions of Newtonian atoms obey classical Newton's second law. The Newton's equations of motion are integrated with a Velocity-Verlet algorithm with a time step of 1×10^{-15} s. Periodic boundary conditions are imposed in the x and y direction for all simulations. Indentation is conducted in displacement control by positioning the spherical indenter, simulated by a repulsive potential, into the (1 1 1) crystallography surface of substrate. The repulsive potential is described as

$$F = AH(r)(R - r)^2, \tag{1}$$

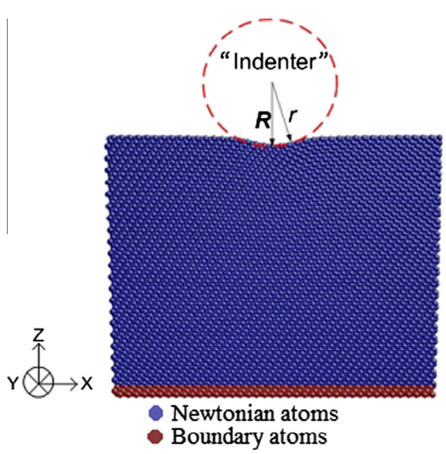

Fig. 1. A schematic 2-D section representation of indentation configuration at $y = 0$. The indenter potential function can be interpreted as a spherical indenter of radius R. The vector connecting the indenter center to any atom is r. In this schematic, r gives the position of a contacted atom; the right-handed coordinate system is shown at the left.

Table 1
Pre-strain vs. corresponding residual stress and normalized by the ideal shear strength of Cu substrate.

Pre-strain (%)	−2	−1	−0.5	0	0.5	1	2
σ_{res} (GPa)	−4.5	−2.15	−1.05	0	0.95	1.9	3.6
0	−0.99	−0.47	−0.23	0	0.21	0.42	0.79

σ_{res} – Residual stress resulted from MD simulation, λ – Relative residual stress, $\lambda = \sigma_{res}/G_{ideal}$, where G_{ideal} the ideal shear strength ($G_{ideal} = 4.56$ GPa) of the Cu (1 1 1) single crystals [11].

where A is force constant with a value of 10 eV A^{-2}, $H(r)$ is a step function, R is the indenter radius, and r is the distance from the atom to the center of the indenter sphere. All the simulations are performed at 0.1 K under the NVT ensemble. To discern defects, colors are assigned to the atom according to a local classification visualized by the centro-symmetry parameter (CSP) [12].

3. Results and discussion

3.1. Load vs. indention depth response under different residual stress state

The load vs. indention depth curves with and without residual stress is plotted in Fig. 2. It can be observed that all the curves well overlap in the elastic regime, and agree with Hertzian contact theory, which relates the applied load P to the indenter depth h as [13]:

$$P = \frac{4}{3}E^{*}R^{1/2}h^{3/2}, \tag{2}$$

where the reduced modulus E^{*} describes the material's elastic response, and could be obtained by fitting from MD simulation results through Eq. (2). All the fit values are similar and displayed in the inset of Fig. 2. Although the fit values have some deviation from the literature value of 190.3 GPa [11] due to crystal anisotropy [14], the results indicate that the residual stresses have little effect on the elastic property of single-crystal copper thin film.

In order further to reveal the onset of plasticity, indentation hardness is investigated. The indentation hardness is normally defined as a mean pressure p_m, as:

$$H = p_m = \frac{P_{max}}{A_c}, \tag{3}$$

where P_{max} is the critical load, at which dislocation nucleation occurs, A_c is the indentation contact area at the critical load. The critical area A_c is evaluated from the projected polygon approach [7], and is shown in Fig. 3. Then, the indentation hardness can be obtained through Eq. (3), and shown in Fig. 4. Starting from the state with no residual stress ($\lambda = 0$ in horizontal-axis in Fig. 4), we can find that the indentation hardness rapidly decreases with the increasing tensile residual stress. We may also find that the indentation hardness increases with the increasing compressive residual stress when it is in a moderate range. This phenomenon is consistent with the experiment and/or finite element simulation results in the literatures [15,16]. However, when compressive residual stress exceeds a certain value ($\lambda = -0.47$ in Fig. 4), the indentation hardness will start to decrease. This is one of interesting findings in this study. From stability point of view this phenomenon can be roughly understood in terms of that the comparable large in-plane compressive residual stress could deteriorate the local surface structure stability when a out-plane contact load is applied. The micro-mechanism for this phenomenon will be discussed in the following subsection.

The initial dislocation nucleation depth under different residual stress levels is also investigated from MD simulation results. The