MOLECULAR DYNAMICS SIMULATION OF INDENTATION ON SILVER COATED COPPER NANOSTRUCTURE

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Abstract
The effect of misfit on the indentation behaviour of silver coated copper multilayer was studied by molecular dynamics simulation. It was found that the misfit bands on interface formed by the mismatch of lattice structure between copper and silver in slip direction [110] and the dislocation band width depended on the mismatched lattice constants of materials. More dislocations were created and glided by indentation, which created a “four-wing flower” structure consisting of pile–up of dislocation at the interface. The size of “flower” depended on the thickness of silver layer. The critical thickness for “flower” was approximately 4nm above which the “flower” disappeared. As the result, deformation mechanisms such as dislocation pile-up, dislocation cross-slip and movement of misfit dislocation were revealed. Only silver atoms in the dislocation pile-up were involved in the creation of the “flower” while the dislocations in copper were glided in slip direction on interface.

INTRODUCTION

Because of the increasingly important thin film technology, the plastic behaviour of geometrically confined systems has been in the focus of recent research, and atomistic methods are being used to study the plastic behaviour. Mechanical deformation, fracture, and friction of solids and thin films pose some of the most interesting computational challenges for atomistic calculations of materials properties. The fundamental goal is to establish a connection between atomic scale processes and measurable mechanical properties of materials. The development of nanoindentation techniques has generated considerable interest in the detailed mechanisms of deformation during indentation at a very small scale. Indentation techniques measure the mechanical properties of thin films and other materials (e.g., elastic constants, yield stress, and hardness) by measuring the force or load on the indenter tip as a function of the tip displacement during indentation and retraction.

Previous atomistic calculations have studied nanoindentation and retraction using the Torsoff potential for simulation of β-SiC with diamond indenter is reported about the dependence of the critical depth and pressure for the elastic-to-plastic transition as a function of indentation velocity, tip size, and work-piece temperature by Noreyan *at al* [1]. Fang and Wu [2] applied empirical potential for simulation on nanoindentation of multilayered films. They used Morse potential for calculation of bond energy with the tight-binding second-moment approximation potential adopted to simulate the interatomic energy of Ni–Ni, Al–Al, and Al–Ni atoms. The Lorentz–Berthelot mixing rule was used to estimate the interatomic Morse potential for C–Al and C–Ni atoms. The rest, about simulation of nanoindentation employed Embedded Atom Method (EAM) [3]. Besides, Interatomic Potential Finite Element Method (IPFEM) is used for 2D simulation of nanoindentation, Van VLiet *et al* [4] used this method to compare MD simulations and verify the slip character of the homogeneously nucleated dislocations predicted by the Λ criterion.

Grain boundaries hinder dislocation activity and create dislocation pile-up, thus making metal harder to deform. Reduction in grain size creates more obstacles to dislocation motion due to strengthening of polycrystals. The increase in yield stress is found to be inversely proportional to square root of the grain size, known as the Hall-Petch relation [5,6]. However strength of nanomaterials does not increase continuously with decreasing grain size, below a critical grain size, experiments have shown that the strength decreases again. This referred to as inverse Hall-Pitch [7,8,9]. Multilayer thin metal films with nanometer layer thickness manifest mechanical behaviour similar to nanocrystaline metals. Operating deformation mechanisms in multilayered thin film are similar to nanocrystalline metals. The volume fraction of grain boundary increases as the layer thickness decreases. A large portion of plastic deformation accumulates in the grain

boundaries and grain boundary driven plasticity dominates over the dislocation slip observed in single crystal thin film [4,10,11]. The interaction of dislocations and interfaces become are controlling parameter of plasticity in such systems. Creation of dislocation at interfaces, transmission of dislocation through interfaces are emission of dislocation from interfaces [12-15].

In this paper, we focus on interaction of dislocations and interface of Cu/Ag multilayer indentation, observation in dislocation split and effect of dislocation pile-up on mechanical properties, also inverse Hall-Pitch relation at nanoscale.

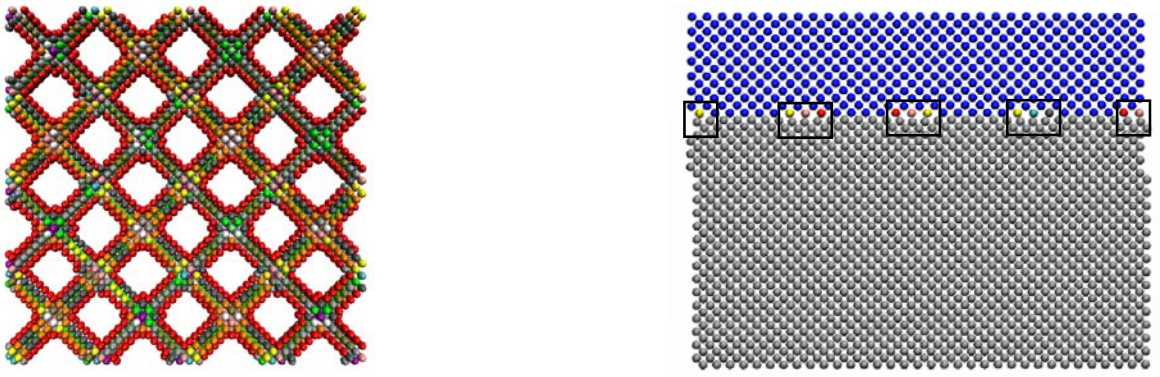
SIMULATION DETAILS

In this work, Embedded Atom Method [3] is employed to simulate nanoindentation of Cu/Ag multilayer thin film, the Jonson's model [16] is applied with the interatomic potentials defined by Wadley *at al* [17]. All simulations of indentation use parallel molecular dynamics program LAMMPS [18] with radius of spherical indenter of 40 Å. The transverse dimensions are approximate 120x120 Å for all models with different coating layers thickness. The bottom layers of atoms are held fixed while periodic boundary conditions are applied for the side surfaces. All models are objected to relaxation for equilibrium state at zero temperature by energy minimization using the conjugate gradient method. Different lattice constants of copper and silver cause a mismatch on the interface between two metals. The extra planes are introduced into the layer with smaller lattice constant, so the creation of edge dislocations on interface is known as misfit. The spacing of misfits depends on lattice constants of two metals, so transverse dimension for models should be common multiple of misfit spacing and are calculated following formula.

$$l = k\beta \frac{a_1 a_2}{|a_1 - a_2|} \tag{1}$$

Where: a_1, a_2 are lattice constants of two metals; k is number of multiple lattice spacing, and is positive integer
β is transverse direction coefficient

The formation of two pairs of full misfit dislocations in interface is got after relaxation (Fig 1). The misfit dislocations on (001) plane of the interface is shown as dislocation network. The lines of network lie along <110> directions, the Burgers vectors of edge dislocation on the network-line are slip vectors on (001) plane, and each dislocation on network has same Burgers vector $\frac{a}{2}[110]$ or $\frac{a}{2}[\bar{1}10]$. Centrosymmetry parameter method [19] is applied for detecting dislocation atoms, only dislocation atoms are plotted (fig 1a) for accelerate displaying of model and easily detecting dislocation region. Dislocation atoms with different centrosymmetry parameters are plotted by different colours.



a **b**

Figure 1 the structure of the relaxed interface, (a) top view of misfit network in interface, (b) (010) cross section

RESULTS AND DISCUSSIONS

Indentation of copper-silver multilayer thin film as also copper and silver thin film are carried out with constant indenter tip velocity of 20 m/s using absolute hardness spherical indenter with radius of 40 Å. The kinetic energy is added into the system by pressing the indenter into surface is dissipated by a Nose-Hoover thermostat set to 0K. The atomic force between each local atoms and indenter is calculated following

$$F(r)=\begin{cases}-k(r-R)^2 & r>R\\ 0 & r<R\end{cases} \tag{2}$$

Where k is the specified force constant, r is distance from the atom to the centre of indenter, and R is radius of indenter.

The force-indentation depth curves are shown in fig 2. It is clear that the copper possesses higher stiffness than silver and silver coated copper. Plastic deformation of silver layer is local deformation near the indented region. This region does not include the atoms on copper layer. Because copper substrate layer is still elastic state (it will be clear next section about dislocations structure), the material properties of Ag coated Cu layer depend on Ag layer and misfit dislocation like grain boundary. The stiffness of Cu/Ag multilayer is even lower than pure silver, it seems that, grain boundary has strongly effect on material properties, and stronger effect for thinner coating layer (20 Å and 30 Å silver layers). On the contrary, weaker effect for thicker coating layer (40 Å and 50 Å silver layers). Moreover, increasing the thickness of coating layer increases stiffness of silver film (fig. 2b), it proves the inverse Hall-Pitch relation at nanoscale.

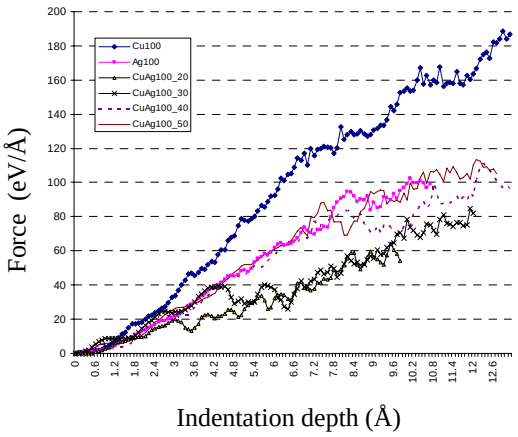


Figure 2 Load-indentation depth curves, (a) Cu, Ag single crystal thin film and Cu coated with Ag(thickness of 20, 30, 40, 50 Å)

Dislocations are produced during nanoindentation into copper with 30 Å silver layer coating are shown in fig 3. In these figure, atoms are plotted if their centrosymmetry parameter exceeds 0.5 Å2 while two surfaces are displayed in point matrix to look inside dislocation structure easily. The red colour is plotted for atoms with centrosymmetry parameter from 0.5 $Å^2$ to 5 $Å^2$ (dislocation) and the yellow colour for stacking fault with the parameter from 5 $Å^2$ to 18 $Å^2$. Pyramid dislocations are created (fig. 3a) and continue to glide down and touch on interface. The pile-up of dislocations (fig. 3c,d) produced "four-wing flower" dislocation structure when dislocation glide up. This dislocation structure is a combination of (111) stacking faults which are bounded by partial dislocations glided up from misfit. The size of the dislocation "flower" is nearly equal to four times of misfit spacing (fig. 3e) for coating layer of 30 Å. While it is only nearly equal to three times for coating layer of 20 Å in fig. 3g. As the result, the size depends on the thickness of coating layer. But the creation of dislocation "flower" only happen with very thin coating layer, the simulations for thicker coating layer than 40 Å can not create the dislocation "flower" however the dislocation pile-up still happens. Dissociation of the edge dislocation into two partial dislocations only happens in silver nearby interface while the edge dislocation in copper still lies in interface (fig. 3h).

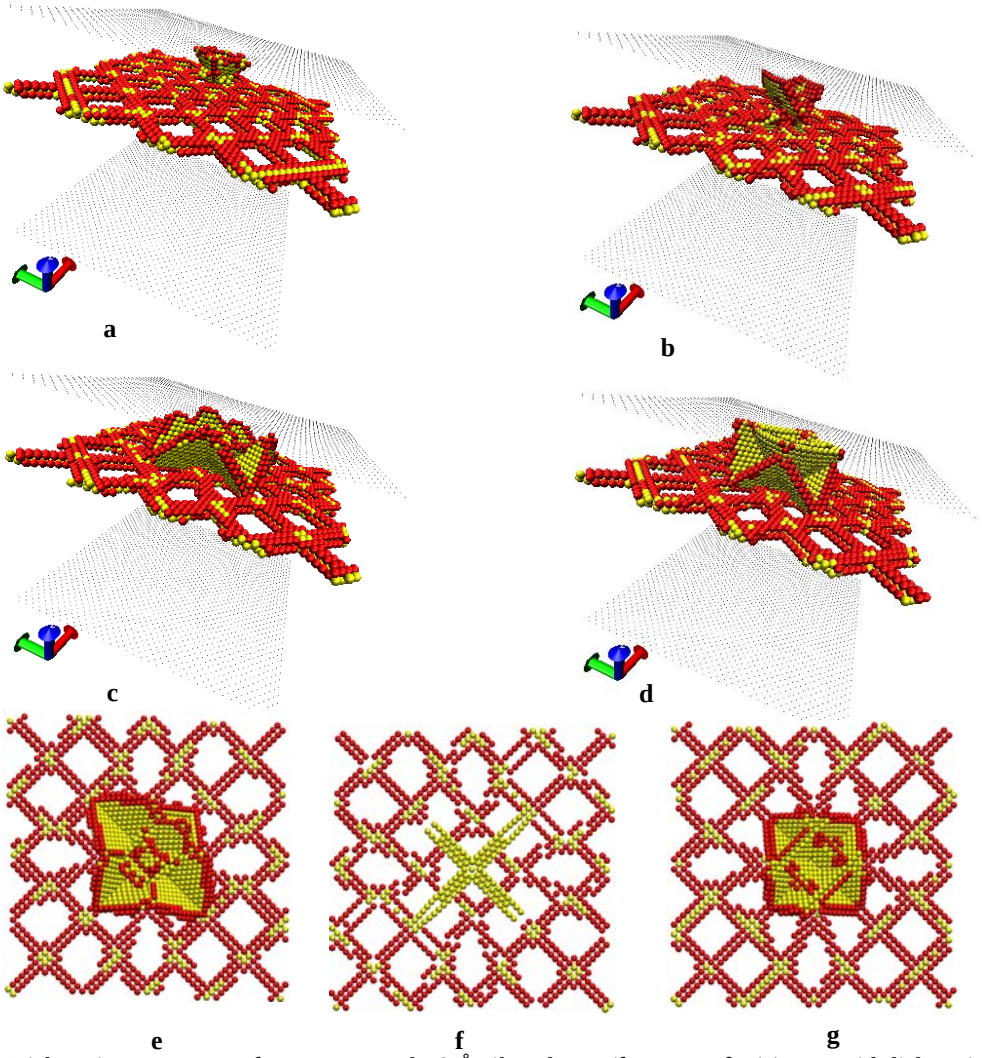


Figure 3 Dislocation structure of copper coated 30 Å silver layer (from a to f), (a) pyramid dislocation at indentation depth of 4 Å; (b) the contact of dislocations on interface at indentation depth of 4.5 Å; (c), (d) movement of dislocations at indentation depth of 5 Å and 5.5 Å, (e) top view of dislocation structure at indentation depth of 5 Å, (f) bottom view of copper dislocation atoms in interface at indentation depth of 5 Å, (g) top view of dislocation structure of copper coated 20 Å silver layer at indentation depth of 3 Å.

Both copper and silver are face-centred cubic, their slip plane is (111) plane [20]. So the misfit dislocation in interface can not glide on this plane (001). When pile-up of pyramid dislocation and misfit dislocation (fig 4) are nucleated for glide up of full dislocation with Burgers vector of $\frac{a}{2}[110]$, which is dissociated into two Shockley partial dislocations (fig 4b). The combination of partial dislocations bound by stacking fault at intersection of two (111) stacking faults creates the Lomer-Cottrell dislocations (fig 4c). To understand the details of dislocation reactions, a schematic representation of fig. 4b is drawn in fig. 4c and Burgers vectors of dislocations on line senses are redrawn in fig. 4d. Burgers vector have to be

perpendicular to line sense and normal vector of stacking fault (slip plane). Conservation of Burgers vectors of the dislocation is analysed in following.

$$\text{At point } A_1\text{: } \frac{a}{2}[\bar{1}10]=\frac{a}{6}[12\bar{1}]+\frac{a}{6}[\bar{2}\bar{1}1]+\frac{a}{3}[\bar{1}10] \tag{3}$$

$$\text{At point } B_1\text{: } \frac{a}{2}[\bar{1}\bar{1}0]=\frac{a}{6}[1\bar{2}1]+\frac{a}{6}[\bar{2}1\bar{1}]+\frac{a}{3}[\bar{1}\bar{1}0] \tag{4}$$

$$\text{At point } C_1\text{: } \frac{a}{2}[1\bar{1}0]=\frac{a}{6}[211]+\frac{a}{6}[\bar{1}\bar{2}\bar{1}]+\frac{a}{3}[1\bar{1}0] \tag{5}$$

$$\text{At point } D_1\text{: } \frac{a}{2}[110]=\frac{a}{6}[\bar{1}21]+\frac{a}{6}[2\bar{1}\bar{1}]+\frac{a}{3}[110] \tag{6}$$

Each edge misfit dislocation splits into two Shockley partial dislocations and a residual dislocation which is left in the interface. These residual dislocations are also Lomer-Cottrell dislocations and they resident in intersection of two (111) stacks at the interface.

Concurrently, the Four Lomer-Cottrell dislocations are formed at the point E_1, F_1, G_1, and H_1 when Shockley partial dislocations meet at intersection of the rest two stacking faults.

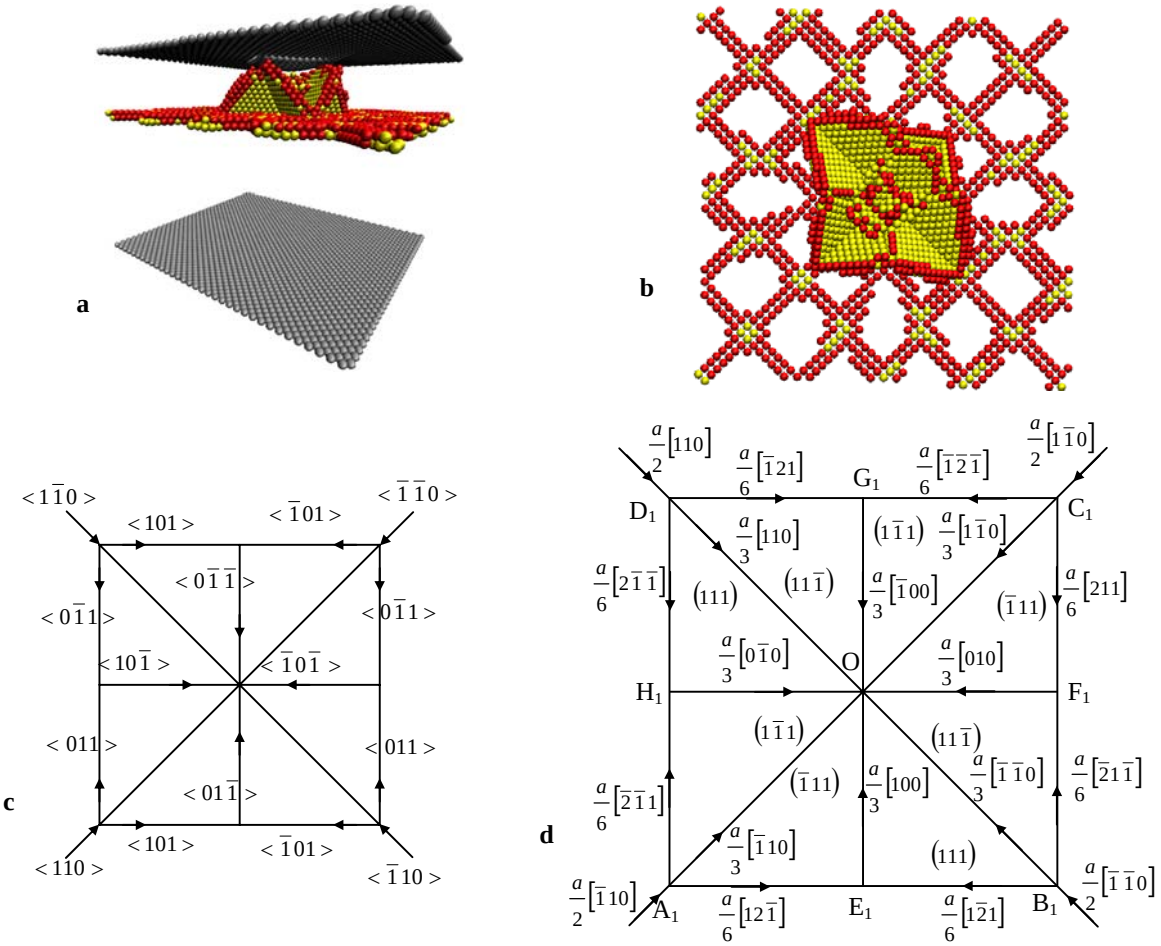


Figure 4 Dislocation structure in copper film with silver coating of 30 Å at indentation depth 5 Å, (a) 3D view, (b) top view of the misfit dislocation in interface, (c) Schematic of misfit dislocation with line senses, (d) Schematic of Burgers vectors

CONCLUSIONS

Nanoindentation of thin copper film/silver substrate multilayer is studied by molecular dynamics simulation method. It is shown that the material properties of copper/silver multilayer depend on thickness of coating layer and misfit dislocation in interface. Creation of dislocation "flower" takes place when the coating layer does not exceeds 40 Å. The glide up of dislocations only happen inside silver layer while the edge dislocation of copper always lies in interface. Burgers vectors of dislocation are observed during dislocations pile-up.

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