

Multiscale Simulation of Surface Defect Influence in Nanoindentation by Quasi-Continuum Method

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Abstract: Microscopic properties of nanocrystal Aluminum thin film have been simulated using the quasicontinuum method in order to study the surface defect influence in nanoindentation. Various distances between the surface defect and indenter have been taken into account. The results show that as the distance between the pit and indenter increases, the nanohardness increases in a wave pattern associated with a cycle of three atoms, which is closely related to the crystal structure of periodic atoms arrangement on {111} atomic close-packed planes of face-centered cubic metal; when the adjacent distance between the pit and indenter is more than 16 atomic spacing, there is almost no effect on nanohardness. In addition, the theoretical formula for the necessary load for elastic-to-plastic transition of Al film has been modified with the initial surface defect size, which may contribute to the investigation of material property with surface defects.

Keywords: nanoindentation; quasicontinuum method; surface defect; multiscale simulation

1. Introduction

Nanoindentation [1], which is relatively simple and effective, has already been a standard technique for evaluating mechanical properties of thin films, widely used in many research fields [2–7]. Recently, a number of relevant scientists have focused on thin films with defects through simulations and experiment [8–11]. Wenshan Yu and Shengping Shen observed the strong effects of the geometry of the nanocavity as a kind of defects in the film during the nanoindentation [12]. So far, surface roughness has become a major concern, which can be treated by the assembly of pits and steps [13,14]. Furthermore, the pitted surface can be usually seen in polycrystalline, microchips, MEMS and nanoindentation technology as one typical kind of defects. Consequently, it is necessary and significant to make an observation on the nature of the pitted surface in nanoindentation. Ni yushan *et al* [15] has already studied nanoindentation of Al thin film compared with surface defect situation and defect free situation by multi-scale simulation, and Zhang Zhongli *et al* [16] has found the delay effect of dislocation nucleation with surface pit in nanoindentation. But the distance effect between the pit and indenter on elastic-plastic transition has not been taken into account, which is especially important to thin film performance in nanoindentation and microchips. Our aim is further to conduct the distance effect of pitted surface on nanohardness by quasicontinuum (QC) method [17], which is an effective way to investigate large scale simulation.

In the present article, fifteen distances of adjacent boundary between surface pit and indenter have been simulated by QC method to investigate the distance effect of pitted surface on nanohardness in nanoindentation. Compared with the nanoindentation on defect-free surface, the distance effect of pitted surface on elastic-plastic transition has been well explored and the theoretical formula of critical load for dislocation emission has been modified with initial surface pit.

2. Methodology

The Quasicontinuum (QC) method [18] is an effective mixed continuum and atomistic approach for simulating the mechanical response, especially in large-scale materials. The Ercolessi-Adams potential, which is one of the EAM potentials [19–22], is applied in this simulation to describe the atomistic behavior. Figure 1 shows the nanoindentation model used in the simulation and the corresponding schematic of local and non-local representative atoms with initial surface pit. The relevant material parameters of model are showed as follows: the crystallographic lattice constant a_1 is 0.4032 nm, one atomic spacing in $[\bar{1} 1 0]$ direction (h_0) is 0.1426 nm, Burgers vector \vec{b} is 0.285 nm, shear modulus μ is 33.14 GPa, Poisson ν is 0.319 and (111) surface energy γ_{111} is 0.869 J/m², which is comparable with the experimental values of 1.14–1.20 J/m². The elastic modules predicted by this potential are $C_{11} = 117.74$ GPa, $C_{12} = 62.06$ GPa, and $C_{44} = 36.67$ GPa. The experimental values extrapolated to $T = 0$ K are $C_{11} = 118.0$ GPa, $C_{12} = 62.4$ GPa, and $C_{44} = 32.5$ GPa [19].

The rectangular indenter is set rigid with its width of 9.32 Å (4 times the lattice constant of Al). It is necessary to note that the indenter size is chosen based on the simulation example in QC method manual and does not affect the behavior in the vicinity of the indenter. The indenter shape is chosen rectangular in this simulation because the boundary of energy field and the distance between the pit and the indenter remain unchanged when driven down into the $(\bar{1}10)$ surface, which is exactly necessary to investigate the distance influence of the pit. The width D and depth H of surface pit are respectively 0.688 nm and 0.730 nm. We take such size value because it is moderate and proper to investigate the distance effect of pitted surface. When the pit is too small, the influence of the pit on the nanohardness is not obvious; when the pit is too large, the variation of nanohardness displays not much sensitive to the distance between the pit and the indenter. In the out-of-plane direction, the thickness of this model is equal to the minimal repeat distance with periodic boundary condition applied. The distance of adjacent boundary between the pit and indenter shows d in Figure 1. Fifteen different distances of adjacent boundary d have been simulated in this paper, which is respectively $1d_0, 2d_0, 3d_0, 4d_0, 5d_0, 6d_0, 7d_0, 8d_0, 9d_0, 10d_0, 11d_0, 12d_0, 13d_0, 17d_0, 21d_0$, where d_0 is 0.2328nm (one atomic lattice spacing in [111] direction). These distance situation are selected in order to make a more comprehensive investigation.

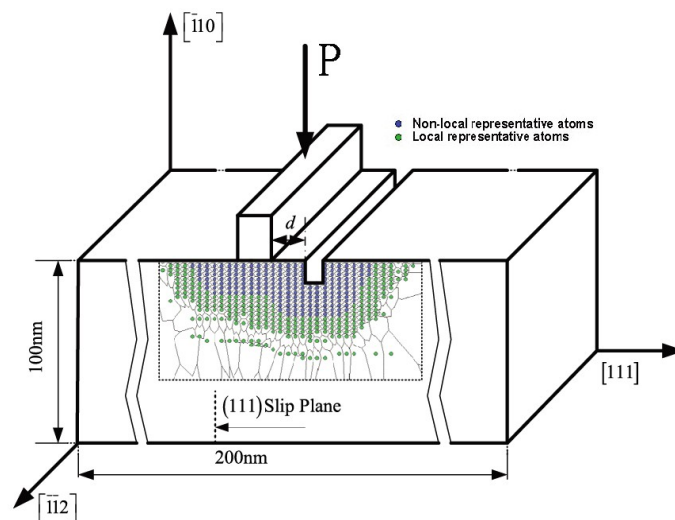


Figure. 1 The schematic illustration of nanoindentation model with a concave defect, where the unusual shapes in local region are not finite elements, they are just the schematic of its specific region that one corresponding representative atom belongs to.

The thin Al film in this simulation is $0.1\mu\text{m}$ in height and $0.2\mu\text{m}$ in width as shown in Figure 1, which is very large in normal atomistic modeling standards with almost 1.3 million atoms or about 4 million degrees of freedom. By contrast, multiscale simulation by QC method uses continuum assumptions for reducing the degrees of freedom and computational demand without losing atomistic required details. At most only 4000 atoms or 12,000 degrees of freedom are treated and such simulation can be run on personal computer in few days.

3. Results and Discussion

3.1. Nanohardness in the Case of no Surface Defect

To make a comparison, a study of nanoindentation on defect-free surface is carried out. The load-displacement curve shown the basic information obtained from nanoindentation simulations on defect-free surface is presented in Figure 2, where Load is expressed by length units of indenter in the out-of-plane direction with its unit N/m. It can be seen from Figure 2 that the load curve gradually increases during initial loading process (OA), which indicates the elastic stage of thin film. The load increases to a maximum value of 15.14 N/m when the load step reaches 0.48 nm at point A. Then the load experiences an abrupt drop that it continually decreases to a minimum value of 7.67 N/m at point B.

To probe the potential mechanism of such abrupt decline of load (AB segment in Figure 2), the atoms snapshot and corresponding out-of-plane displacements experienced by the atoms are investigated. Figure 3 shows the atoms structure at the step of 0.48 nm and 0.50 nm , which are respectively corresponding point A and point B in Figure 2.

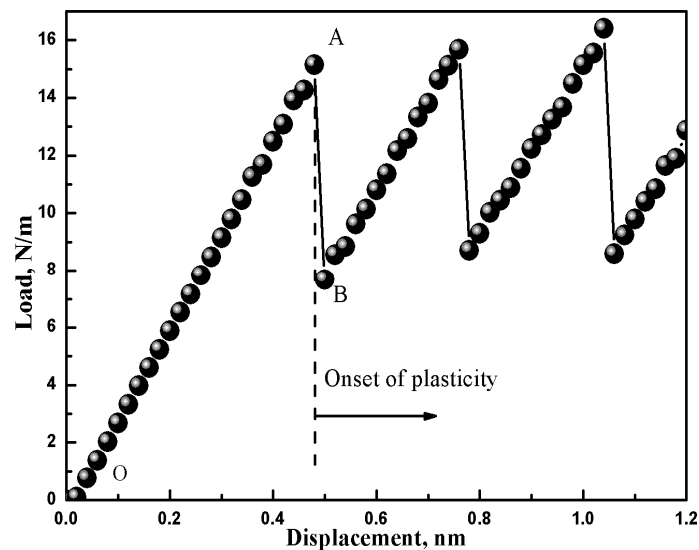


Figure 2. Load-displacement curves for nanoindentation on Al film without surface pit.

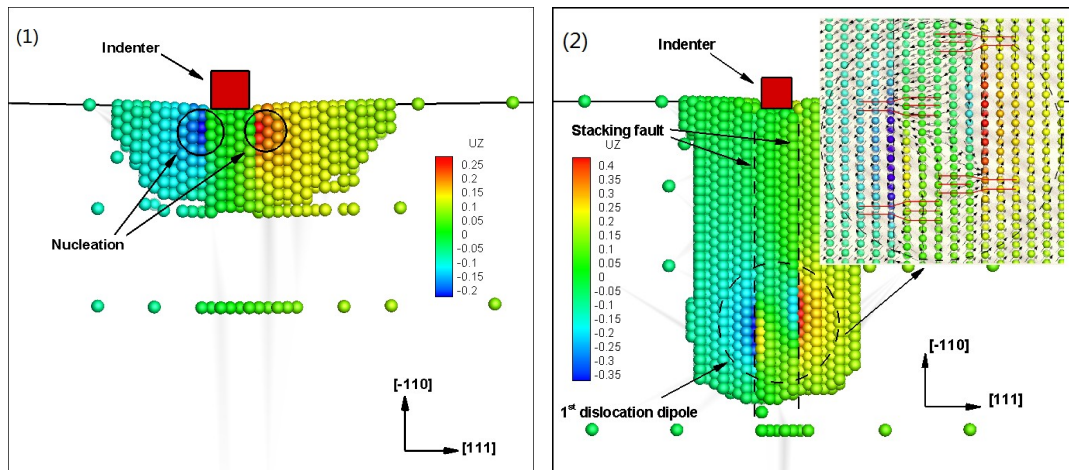


Figure 3. Snapshot of atoms under indenter and corresponding out-of-plane displacement plot, where UZ is atom displacement at out-of-plane. (1) point A in Figure 3 (dislocation nucleation); (2) point B in Figure 3 (dislocation emission).

The conclusion can be drawn from Figure 3 that the load reaches the critical value for dislocation emission at point A, which indicates the beginning of plastic deformation stage. Then two Shockley partial dislocations are emitted at point B. The hardness is given by the equation [23]: $H = \frac{P_{\max}}{A}$, where P_{\max} is the maximum load and A is the indentation area, the nanohardness of Al thin film is 16.24GPa with no surface defect.

In this simulation, the indenter width is 0.932nm, the yield load is about 15.14 N/m, and this is smaller than the value 24.7 N/m obtained in the research of Tadmor and Miller [24] with defect-free surface, where the indentation size is 2.5 nm. It is reasonable that a reduced width of indenter will make significant decrease in yield load, which is consistent with a published observation [25].

3.2. Nanohardness with Various Distances between Surface Defect and Indenter

Figure 4 shows each nanohardness value in the case of nanoindentation on the thin film surface with and without surface pit. It indicates that the nanohardness of pitted surface is reduced, compared with the one of no surface defect. This is reasonable because the atomic structure of thin film has been destroyed by the surface pit. Further, when the distance of adjacent boundary between the pit and indenter d respectively equals $1d_0$, $2d_0$, $3d_0$, $4d_0$, $5d_0$, $6d_0$, $7d_0$, $8d_0$, $9d_0$, $10d_0$, $11d_0$, $12d_0$, $13d_0$, $17d_0$, $21d_0$, the nanohardness curve rises up in a wave pattern and finally tends towards the nanohardness value of no surface defect.

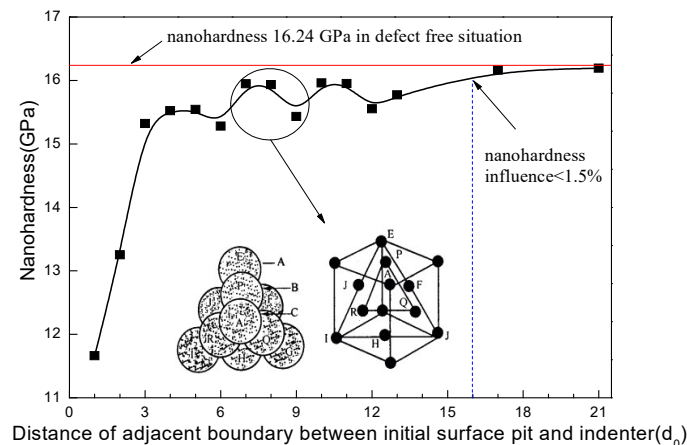


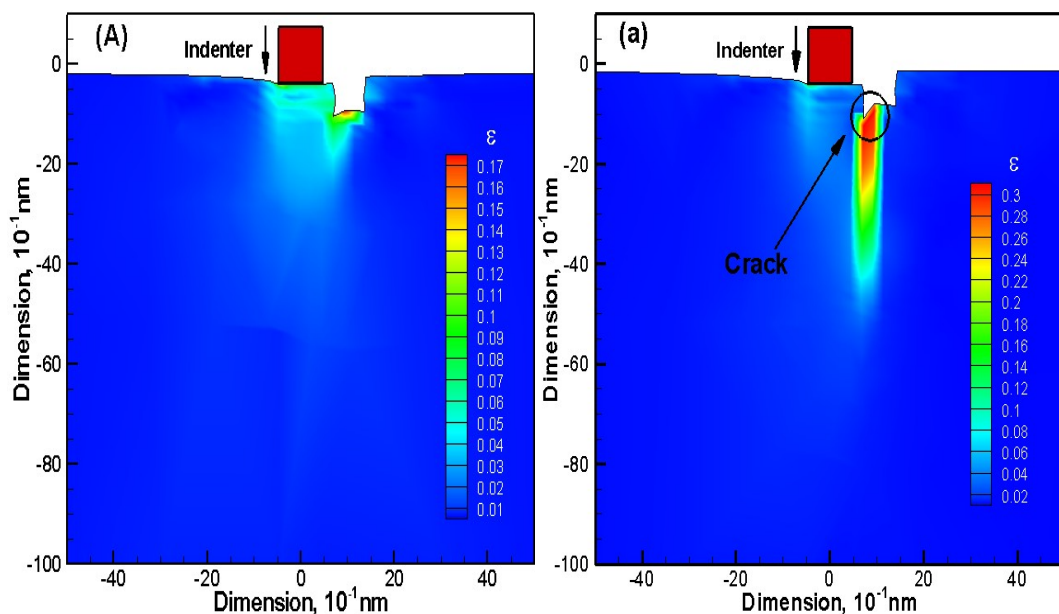
Figure 4. Nanohardness *vs* distance of adjacent boundary between the pit and indenter.

Through a further investigation, the change law of nanohardness is discovered that such wave pattern is associated with a cycle of three atoms (donated by circle in Figure 4), which is closely related to the crystal structure of periodic atoms arrangement. The scientific definition of a "crystal" is based on the microscopic arrangement of atoms inside it, called the crystal structure, where the atoms form a periodic arrangement [26]. Moreover, different stacking pattern of atoms has different gaps between adjacent atoms, which make a great influence on the performance of metal. In this simulation, such periodic arrangement of atoms is "ABCABC" on {111} atomic close-packed planes of face-centered cubic metal (as shown in Figure 4 at the illustration). According to this simulation, when the simulation distance d increases each three atoms spacing on [111] direction, one cycle of atoms arrangement in "ABCABC" pattern is exactly finished. That's why the nanohardness curve increases in a wave pattern associated with a cycle of three atoms.

In order to figure out the spatial extent of influence of surface pit on nanohardness, a further discussion is carried out. As Figure 4 shows, when the distance between the pit and indenter increases, the influence of nanohardness compared with the nanohardness of no surface defect (16.24 GPa) is running low. If the influence of nanohardness is smaller than 1.5%, it can be considered that the surface pit almost doesn't affect the nanohardness. According to this simulation, when the distance (d) is more than 16 atomic spacing, there is almost no effect on nanohardness (as shown in Figure 4). It can be predicted that different material has such different critical value, which has significant meanings to the size design of thin layers in nanoindentation or microchips with the hardness guarantee.

However, the first three atoms, respectively $d = 1d_0$ 、 $d = 2d_0$ 、 $d = 3d_0$ distance, do not match the wave pattern. To explain such special phenomenon, atomic structure and corresponding strain distribution of Al crystal are probed.

Figure 5 shows Von Mises strain distribution of crack propagation and a comparison of strain before and after the crack when the distance d respectively equals $1d_0$ 、 $2d_0$ and $3d_0$. It shows that when the distance d equals $1d_0$ and $2d_0$, there appears a crack phenomenon at the left side of surface pit, which directly induces serious damage to the structure of materials (as shown in Figure 5 (A), (a); Figure 6 (B), (b)). But when the distance d equals $3d_0$, there is no crack (as shown in Figure 5 (C), (c)). Consequently, when the distance d equals $1d_0$ and $2d_0$, the nanohardness is greatly reduced. That is to say, the first three atoms in nanohardness curve as shown in Figure 4 will not match the wave pattern associated with a cycle of three atoms.



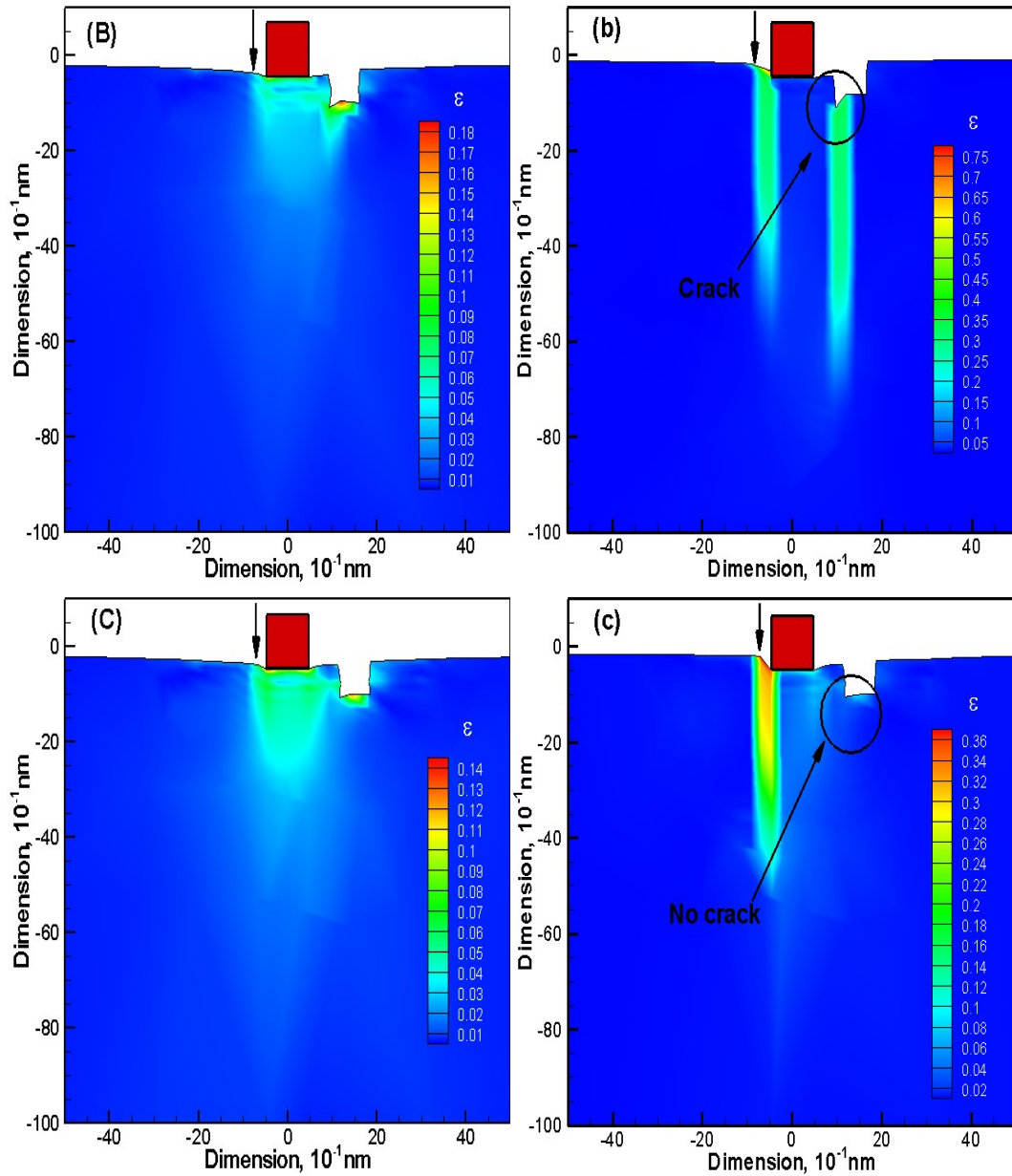


Figure 5. Von Mises strain distribution of crack propagation. (A) $d = 1d_0$ at the load step of the indenter 0.38 nm; (a) $d = 1d_0$ at the load step of the indenter 0.4 nm; (B) $d = 2d_0$ at the load step of the indenter 0.44 nm; (b) $d = 2d_0$ at the load step of the indenter 0.46 nm; (C) $d = 3d_0$ at the load step of the indenter 0.46 nm; (c) $d = 3d_0$ at the load step of the indenter 0.48 nm.

3.3. Formula Modification of Necessary Load for Elastic-to-Plastic Transition

It is known that the influence of surface pit on nanohardness is actually the influence of dislocation nucleation and emission which is affected by surface pit. In order to make a further study on the nanohardness calculation in the case of pitted surface, the calculation formula of necessary load for elastic-to-plastic transition is discussed and modified based on the model with no surface defect.

According to the critical load for dislocation emission, which is applied by Tadmor [24], the calculation formula of critical load for dislocation emission is displayed as following:

$$P_{cr} = \frac{\mu b}{4\pi(1-\nu)} \ln \frac{32h(h+2a)a^2}{b^4} + 2\gamma_{111} + \frac{1}{2}kb \quad (1)$$

where P_{cr} is the critical load value at the onset of dislocation emission, k is the slope of elastic stage in the load-displacement curve, h is the depth of dislocation dipole when it is emitted, a is the half width of indenter, γ_{111} is the energy of (111) surface of Al crystal.

In order to make a more reasonable investigation, the data of simulation that $d=1d_0$ and $d=2d_0$ is not taken into account because of crack.

Table 1. The comparison of critical load between QC method and dislocation theory.

Distance (d_0)	QC data (N/m)	Theory Load (N/m)	Data Difference (N/m)
3	14.28	18.02	3.75
4	14.46	17.29	2.83
5	14.48	17.88	3.39
6	14.24	17.41	3.15
7	14.86	17.96	3.14
8	14.85	17.65	2.83
9	14.38	17.92	3.07
10	14.87	17.87	3.49
11	14.86	18.03	3.16
12	14.49	17.56	2.70
13	14.70	17.99	3.50
17	15.06	18.04	3.34
21	15.09	18.17	3.11

Table 1 shows the comparison of critical load between the QC method and dislocation theory, where “QC data” means the data of critical load in this simulation, and “theory load” means the data of critical load using equation (1). It shows that in the case of every distance (d) simulation, the difference value between QC method and dislocation theory is changed frequently. According to the phenomenon discussed above that the nanohardness is periodically changed in a circle of three atoms, the critical load for dislocation emission is also in such periodicity. Consequently, the correction term (set as Δ) can be separated into two parts:

$$\Delta = A(d) + B \cdot \sin(d) \quad (2)$$

where $A(d)$ is the correction part for the hardness decrease because of the surface pit and $B \cdot \sin(d)$ is just for the periodic change of atoms arrangement. It is well known that when the pit size (D , H as shown in Figure 1) is bigger, the value of critical load of dislocation emission is smaller. So it is

reasonable to apply $\frac{D}{a_1} \cdot \frac{H}{a_1}$ (dividing by the crystallographic lattice constant can significantly make it dimensionless, which has already been demonstrated reasonable in published article [27]) to express the size influence of surface pit. When the surface pit is infinitely far away from the indenter, the influence on nanohardness can be ignored. And if the pit size increases, the correction term changes more slowly with the distance variation. So it is reasonable to apply $\ln(1 + (\frac{d_0}{d})^{\frac{d_0 \cdot h_0}{D \cdot H}})$ to express the distance effect of surface pit. Furthermore, the affection of surface pit is closely related to the material property such as Burgers vector \vec{b} , shear modulus μ and Poisson ν . According to equation (1), it is reasonable to apply $\frac{\mu b}{4\pi(1-\nu)}$ to express the influence of material property. In addition, on {111} atomic close-packed planes of face-centered cubic metal, the periodic atoms arrangement is "ABCABC". So the periodicity is three atoms. That is to say, it is reasonable to apply $\text{Sin}(\frac{2\pi}{3d_0} \cdot d + \varphi)$ to express the periodicity of atoms arrangement. Considering that the unit of correction term (Δ) is N/m, and according to the discussion above, the correction term can be defined as following:

$$\Delta = \alpha \cdot \frac{\mu b D H}{4\pi(1-\nu)a_1^2} \ln(1 + (\frac{d_0}{d})^{\frac{d_0 \cdot h_0}{D \cdot H}}) + \beta \cdot \frac{\mu b}{4\pi(1-\nu)} \cdot \text{Sin}(\frac{2\pi}{3d_0} \cdot d + \varphi) \quad (3)$$

where α 、 β 、 φ are three constants that need to be optimized. According to the simulation data in Table 1, these three constants α 、 β and φ are approximately $\frac{3}{2}$ 、 $\frac{2}{15}$ 、 $-\frac{\pi}{3}$ respectively. So the theoretical formula for necessary load of the first dislocation emission of Al film has been modified with initial surface pit as follows:

$$P_{cr}^* = \frac{\mu b}{4\pi(1-\nu)} \ln \frac{32h(h+2a)a^2}{b^4} + 2\gamma_{111} + \frac{1}{2}kb - \frac{3\mu b D H}{8\pi(1-\nu)a_1^2} \ln(1 + (\frac{d_0}{d})^{\frac{d_0 \cdot h_0}{D \cdot H}}) - \frac{\mu b}{30\pi(1-\nu)} \cdot \text{Sin}(\frac{2\pi}{3d_0} \cdot d - \frac{\pi}{3}) \quad (4)$$

Figure 6 shows the comparison of the necessary load for elastic-to-plastic transition of Al thin film with various distances between the pit and the indenter calculated by the theoretical formula before and after modification. Though there is no parameter d in the unmodified dislocation theory (eq.1), the curve with blocks are calculated by the each depth h corresponding each distance case of surface pit and indenter in this simulation. It shows that the simulation QC data is closer to the theoretical results which are calculated by the equation (4) after modification. That is to say, such modification to the theoretical formula is reasonable and efficient that the pit size and the distance between the pit and indenter have both been taken into account.

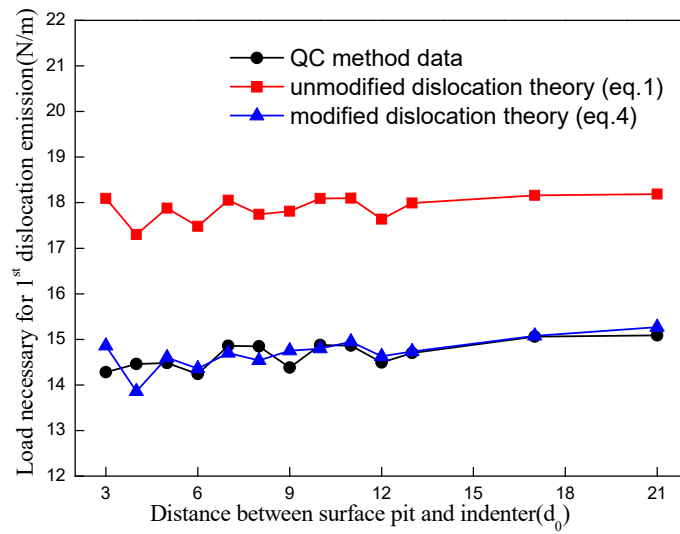


Figure 6. The comparison of the necessary load for elastic-to-plastic transition of Al thin film with various distances between the pit and the indenter calculated by the theoretical formula before and after modification.

This modified formula has well performed the decreasing trend of nanohardness as the distance between the pit and indenter increases. Such trend is greatly agreed with the experimental results of surface step with various distances [14]. Moreover, this modification may contribute to the investigation of the material property influenced by the surface defects, particularly in nanoindentation, MEMS and microchips.

4. Conclusion

In this paper, the QC method is employed to investigate the distance effect of the pitted surface on elastic-plastic transition. Compared with the nanoindentation on defect-free surface, fifteen various distances of adjacent boundary between the pit and indenter are taken into account. The conclusion can be drawn as follows:

- i. The pitted surface plays a great role in the emission of dislocation that it raises significant reduction on nanohardness, compared with defect-free situation.
- ii. As the distance between the pit and indenter increases, nanohardness increases in a wave pattern associated with a cycle of three atoms, which is closely related to periodic atoms arrangement on {111} atomic close-packed planes of face-centered cubic metal; when the adjacent distance between the pit and indenter is more than 16 atomic spacing, there is almost no effect on nanohardness, suggesting that each material has such critical value.
- iii. The theoretical formula for necessary load of the elastic-plastic transition of Al film has been reasonably and efficiently modified with initial surface pit. This modified formula has well performed the decreasing trend of nanohardness as the distance between the pit and indenter increases, and such trend is greatly agreed with the experimental results of surface step with various distances. Such modification may contribute to the investigation of material property with surface defects, particularly in nanoindentation and microchips.

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