Stress at the initial stage of growth for Lennard−Jones films

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Molecular dynamics simulation was used to study the influence of a relative size of adsorbed and substrate atoms on the stress of growing films. Atoms in the system interact via the Lennard−Jones potential. The simulations were performed at a fixed value of systems temperature. The relative size of deposited atoms was changed in the range from 0.7 to 1.2, relatively to the size of substrate atoms. Proposed modeling allows to explain the behaviour of mean biaxial stress for systems with different sizes of adsorbed atoms and a substrate. For considered systems, significant changes in mean biaxial stress have only first three monolayers.

Keywords: film growth, stress, molecular dynamics simulation.

1. Introduction

One of the parameters influencing the stress in thin film systems is a mismatch of the lattice constants between the substrate and films. It is very important to predict the stress evolution during deposition. At the early stage of growth, when the islands of deposited material are created on the surface, the size of adatoms is especially important. Main of parameters, which determined the behaviour of adsorbed films, are the strength of interatomic interaction and system temperature. In experimental cases, the interaction and temperature are usually constants. Then, the relative size of atoms in the system has a significant role.

Recently, an increasing interest in investigations into stresses in thin films, by both experimental [1, 2] and theoretical [3−5], has been observed. Most of the publications were devoted to the modeling of metallic thin films grown by the Volmer−Weber mode. Obviously, due to complex microstructure changes during deposition, all of the proposed modeling studies have been based on simplified mechanics.

In this paper, a molecular dynamics simulation was used to study the influence of the relative size of adsorbed and substrate atoms on the stress of growing films. The simulations were performed at a fixed value of temperature of systems.

2. Model

The results presented in this paper are obtained from molecular dynamics simulations based on the method described in the work [6]. The interactions between the atoms themselves in the film and between atoms in the film and in the surface are assumed to be pairwise additive and the pair potential is represented by a truncated $(12, 6)$ Lennard–Jones potential [7]. The simulations are carried out in a rectangular box of dimensions $L_x \times L_y \times L_z$ $(20 \times 20 \times 30)$ and periodic boundary conditions are applied in the x and y directions. At the bottom of the box, an unmovable solid wall is placed as a substrate. This substrate is assumed to be the (100) plane of an *fcc* regular crystal characterized by the unit vectors a_1 and a_2 of the same length a. All distances and lengths are expressed in the a units and the energy parameter ε is the unit of energy. The particles are created at random (x, y) positions and initially directed perpendicular to the bottom surface. The deposition process consists in repeating the sequence of creating a single atom and equilibrating it at a selected temperature.

The deposition rate V of atoms was given by the number of incident atoms per number of time steps $(V = 1/100)$ and $E_k = 50/\varepsilon$. The reduced temperature $T^* = 0.1$. The time step used for solving Newton's equation of motion is $\Delta t = 0.005$ ps. The temperature during the simulation was kept fixed by periodical velocity rescaling. In our algorithm of simulation, we rescaled velocity every 5–10 time steps.

We assumed that the relative size of deposited atoms σ_r was changed in the range from 0.7 to 1.2, relatively to the size of substrate atoms, $\sigma_r = \sigma_f/\sigma_s$, where σ_f is the size of adatoms and σ_s is the size of substrate atoms. Therefore, in that case, the cut-off distance $r_{\max} = 3.5\sigma_f$ for both types of interactions.

For atomic systems, the stress is determined by the position of atoms, resulting from interatomic potential. For the stress calculation, we used the virial stress based on a generalization of the virial theorem of Clausius [8, 9] for the gas pressure.

3. Results

Figure 1 shows the results of the mean biaxial stress versus the number of deposited atoms for different σ_r. Obtained curves of stress evolution are complicated. But it can be distinguished four types of behaviour.

Figure 1**a** shows that with an increase in σ_r, the tensile stress increases and the compressive stress decreases, but these changes are small. For $\sigma_r \in (0.80, 0.87)$, the similar behaviour can be observed – the compressive stress increases and the tensile stress decreases, as it is shown in Fig. 1**b**. The results presented in Fig. 1**c** show
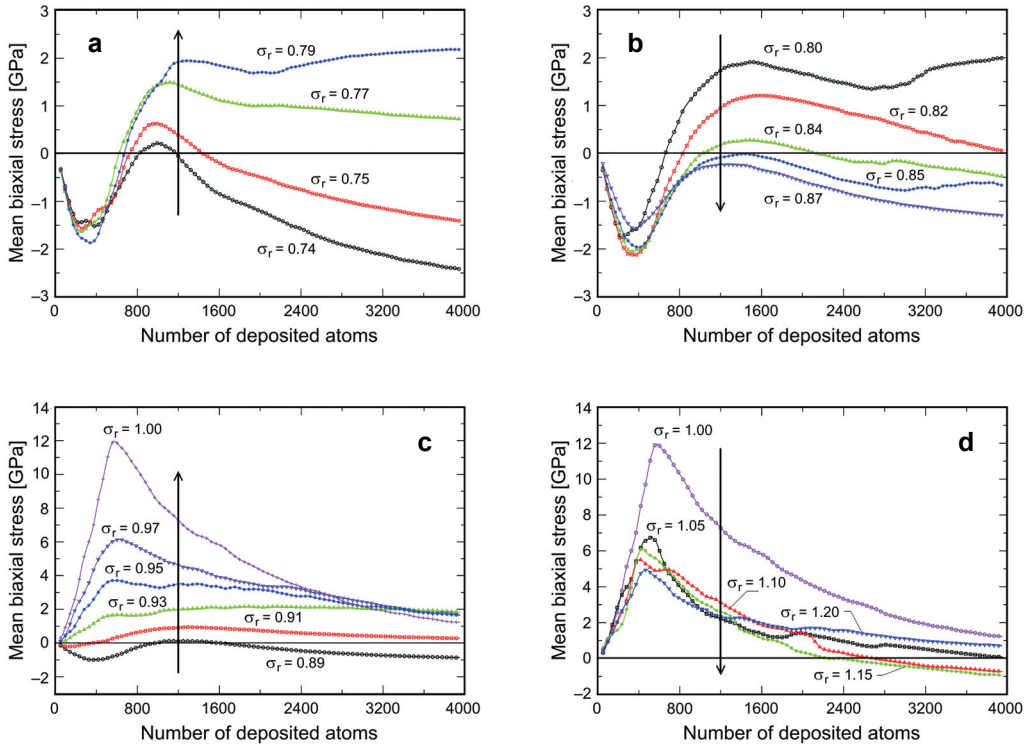
Fig. 1. Mean biaxial stress evolution during simulation versus number of deposited atoms for different sizes of deposited atoms; $V = 1/100$, $T^* = 0.1$.

that the compressive stress disappears and the tensile stress rapidly grows. For $\sigma_r > 1.0$ the compressive stress is not observed and the tensile stress decreases.

Our earlier investigations [6] demonstrated that several monolayers have an influence on the mean biaxial stress in a film. For all considered systems, first three monolayers only had a significant contribution. In order to understand the evolution in stress in such films, we performed a simulation of systems consisting of a substrate and one monolayer of deposited atoms. In this simulation, the deposited atoms were equilibrated. Then, we calculated the mean biaxial stress. Next, we put the monolayer and again it was equilibrated. After equilibrating the obtained films, we put the third monolayer and we equilibrated it, and calculated the mean biaxial stress. Each equilibration was performed for a sufficient number of time steps. The criterion for finishing the equilibration was the energy fluctuation in the system.

The obtained results are shown in Fig. 2. Significant changes in the mean biaxial stress with an increase in σ_r were observed. For systems consisting of one monolayer, we observed a maximum of compressive stress at $\sigma_r \approx 0.80$. Moreover, the mean biaxial stress is zero for $\sigma_r \approx 0.91$. It is correlated with disappearing of the compressive
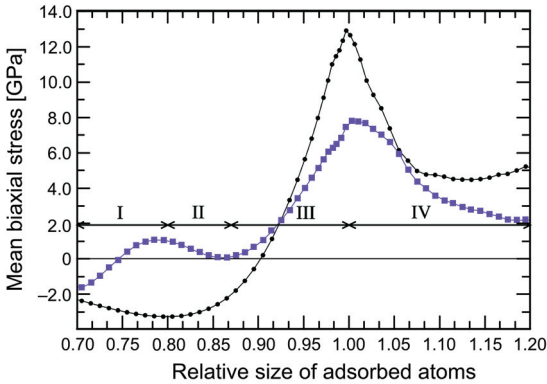
Fig. 2. Mean biaxial stress evolution for one and three monolayers for different relative sizes of deposited atoms σ_r: ● – one monolayer; ■ – three monolayers.

stress at the initial stage of growth. The results for three monolayers are quite different. For $\sigma_r < 0.91$ we observed such kind of fluctuation on the mean biaxial stress with an increase in σ_r. This fluctuation are connected with existing the second and third monolayers. Using these results, we can predict the behaviour of the maximum tensile stress in deposited films.

Particularly, for $\sigma_r < 0.80$ (I) we observe an increase in the mean biaxial stress. In Fig. 1**a**, these changes are correlated with the result of the tensile stress. Next, in the period II, stress decreases. These changes are again correlated with the results of the tensile stress in Fig. 1**b**. In Figure 1**c**, when $0.87 < \sigma_r < 1.0$, the tensile stress again increases. In Figure 1**d**, for the last period (IV), the tensile stress decreases with an increase in σ_r.

4. Conclusions

In summary, our simulations demonstrate that changes in the evolution of stress can be predicted using the simple simulation. Basing on the results presented in Fig. 2, we may explain the behaviour of the mean biaxial stress for the systems with different sizes of adsorbed atoms and a substrate. For the considered systems, a significant change in the mean biaxial stress has only first three monolayers. The required knowledge is the relative size of adsorbed atoms. Of course, the proposed method is very simple and limited.

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