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Molecular dynamics simulation of stress and grain evolution

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A three-dimensional molecular dynamics simulation is carried out to study the evolution of grains and stresses during the deposition of atoms on the (100) plane of a fcc regular crystal, using cubic system with x-y periodic boundary conditions. At the bottom an atomic surface and at the top a reflecting wall are assumed. Atoms in the system interact via the Lennard–Jones potential. During simulation the films grow according the Volmer–Weber mode and exhibit specific shape of the stress curves. When the film becomes continuous, the stress during the growth possess a maximum value, but later new grain boundaries are formed. Individual atoms in the grain boundaries generate compressive stress in the films.

Keywords: film growth; stress; grain; molecular dynamics simulation

1 Introduction

Stresses are usually generated during the process of deposition of thin films, irrespective of the deposition technique applied, and one can influence the reliability of evaluation of the properties of the deposited system by applying appropriate techniques [1]. The stress often leads to unfavorable consequences for defective

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functioning or even to breakdowns of thin-film elements. Therefore, recognition of phenomena that are mainly responsible for the stress in thin metal films is of significance.

Recently, there has been observed an increasing interest in the investigation of stresses in thin films prepared by different techniques [2–10]. Much of publications were devoted to the modeling of metallic thin film grown by Volmer–Weber mode. These modeling studies explain the behavior of experimental stress-thickness curve by taking into account processes of nucleation and growth of islands, coalescence and post-coalescence film growth of islands. Obviously, due to complex microstructure changes during deposition, all of the proposed modeling studies have been based on simplified mechanics.

Computer simulations have an enormous potential to provide better understanding of the growth and stress generation of thin films as well as to mimic their real deposition processes. Recently, several authors have employed molecular dynamics simulation to understand the behavior of stresses in films [11–13]. In particular, Zhang et al. [11] used molecular dynamics simulations to study the stress generation mechanism in thin films grown by ion-beam deposition, whereas Kalyanasundaram et al. [12] presented results of stress evolution during ion bombardment in silicon.

This work is addressed to the general aspects of stress evolution and its relationship with the evolution for grains during the deposition of thin films at different temperatures. In contrast to the previous investigation, we consider here interactions between particles in continuous space only, using three-dimensional molecular dynamics simulation to model thin film deposition onto an initially flat surface. We study the stress and grains evolution during growth thin films at different temperatures.

1 In three-dimensional molecular dynamics simulations of thin-film growth applied
 2 in this work the trajectories, of individual atoms in the computational cell are
 3 obtained by integrating Newton's classical equation of motion. The integration is
 4 performed over finite time steps using a fifth-order predictor–corrector method [14].
 5
 6 The interactions between the particles themselves in the film and between particles
 7 in the film and in the surface are assumed to be pairwise additive and the pair
 8 potential is represented by a truncated (12,6) Lennard–Jones potential:
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$$U(r) = \begin{cases} 4\varepsilon[(\sigma/r)^{12} - (\sigma/r)^6] & r \leq r_{\max} \\ 0 & r > r_{\max} \end{cases}, \quad (1)$$

15 where r is the distance between the particles, ε is the strength of the interactions,
 16 and r_{\max} is the cut-off distance, equal to about 3.5σ for both type of interactions.
 17 We also assume that the size σ of the atoms is constant i.e. $\sigma = 1$, and $\varepsilon = 1$.
 18 The simulations are carried out in a rectangular box of dimensions $L_x \times L_y \times L_z$
 19 and periodic boundary conditions are applied in the x and y directions. At the
 20 bottom of the box, an unmovable solid wall is placed as a substrate. This substrate
 21 contains 3840 atoms forming 15 layers. This substrate is assumed to be the (100)
 22 plane of an fcc regular crystal characterized by the unit vectors a_1 and a_2 of the
 23 same length $|a_1| = |a_2| = a$. Throughout this paper, all distances and lengths are
 24 expressed in the units of a , and the energy parameter ε of the potential (1) is the
 25 unit of energy.
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47 To prevent the atoms from leaving the simulation box, a reflecting wall is placed
 48 at its top. The source that creates particles in the system is kept at a sufficient dis-
 49 tance away from the bottom surface. This distance is equal to $45a$ in our case. The
 50 particles are created at random (x, y) positions and initially directed perpendicular
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to the bottom surface. The simulation is divided into two stages: deposition and equilibration. The deposition process consists of repeating sequence of creating a single atom and equilibrating it at a selected temperature. Finally, the obtained system is equilibrated. The averages of the interesting quantities are accumulated after each 60 time steps and the number of steps used for the equilibration is 10^5 .

The reduced time is defined as usual [14], $t_0 = (m\sigma^2/\varepsilon)^{1/2}$, where m is the mass of the atom. The time step used for solving Newton's equation of motion is $\Delta t = 0.005t_0$. Hereafter the time is expressed as a number of the time steps. 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. The reduced temperature $T^* = k_B T/\varepsilon$, where k_B is the Boltzmann constant and T is the temperature in Kelvin. The deposition rate of atoms was given by the number of incident atoms per number of time steps and always the energy of deposition $E_k^* = 50/\varepsilon$.

The commonly used definition of stress for atomic systems is the virial stress. This definition is based on a generalization of the virial theorem of Clausius [15] for the gas pressure. According to this definition, the average virial stress tensor (S) over an effective volume is the sum of contributions from all the atoms in the domain of the volume Ω :

$$S = \frac{1}{\Omega} \sum_i^N \left(m_i v_i \otimes v_i + \frac{1}{2} \sum_{j \neq i} r_{ij} \otimes \frac{\partial U(r_{ij})}{\partial r_{ij}} \right), \quad (2)$$

where m_i is the mass of the atom i , v_i is its velocity, $r_{ij} = r_i - r_j$ is the spatial vector between atoms i and j , N is the total number of atoms in the domain, and \otimes denotes the tensor product of the two vectors. The sign convention adopted here for the force is positive for attraction and negative for repulsion. Accordingly, a positive stress indicates expansion whereas a negative stress indicates compression.

energy of atoms, while the second part comes from interatomic forces. For solids, the kinetic energy term is usually small when compared with the interatomic force term and is ignored. The stress is a (3×3) tensor, which can be represented in the diagonal form:

$$\bar{S} \equiv \begin{pmatrix} S_{xx} & 0 & 0 \\ 0 & S_{yy} & 0 \\ 0 & 0 & S_{zz} \end{pmatrix}, \quad (3)$$

where S_{xx} , S_{yy} and S_{zz} are stresses in the x , y and z directions. The film stress was calculated when the system equilibrated after each atom deposition.

3 Results and Discussion

We performed a series of molecular dynamics simulation of growing films on solid surfaces. During the simulation process, the temperature T^* and the rate of deposition V were changed.

An examples of the dependence of the biaxial stress [$S_{xy} = (S_{xx} + S_{yy})/2$] and the normal stress [S_{zz}] on the number of deposited atoms for the rate $V = 1/60$ are shown in Fig.1. For all temperatures one can observe significant stress evolution during the simulations. Generally, the shapes of the curves of biaxial and normal stress evolution are similar for different temperatures. We found that the changes in the biaxial stress are higher than those in the normal stress. In the initial stage, the biaxial stress increases with an increase in the number of deposited atoms in the tensile direction. Thereafter, a maximum value of the stress is achieved when the number of deposited atoms approaches about 500. This number is characteristic and is independent of the substrate temperature. After attaining maximum value, the biaxial stress changes toward compressive direction and keeps the same trend until

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the end of deposition. For temperatures $T^* = 0.3, 0.5$ and 0.7 one finds that, with the increase in the temperature, the changes are slower and the stress changes from tensile to compressive. The maximum value of the biaxial stress strongly depends on the temperature and decreases with increasing temperature of substrate. Such an evolution of the biaxial stress is often associated with changes in the grain structure of thin films [16].

In contrast to the biaxial stress, the stress in the normal direction is relatively less sensitive to the number of deposited atoms and its value is lower than that of the biaxial stress. In this case, one does not observe stress in the normal direction in the initial stage. However, when the biaxial stress attains the maximum value one also observes an increase in the normal stress. Thereafter, with an increase in deposition, the value of the normal stress decreases and ultimately approaches zero.

In order to analyze the grain structure of the films, we calculated some important parameters connected with the grain structure. Different criteria are available for determining whether an atom belongs to a grain [17]. One option is to consider the energy that binds an atom to other atoms already in the grain. An alternative method requiring less computation (that for attractive pair potentials is essentially the same) is based on the criterion of the interatomic distance. If atom i is already in the grain, atom j will also be included if $r_{ij} < r_d$, where r_d is the chosen threshold separation. In our simulation, this definition was applied. In the calculations we used $r_d = 1.06\sigma$. This value of r_d gives good value of grain properties.

Fig. 2 shows the dependence of average grain size ($\langle s_c \rangle$) on the number of deposited atom for the rate $1/60$ at different temperatures. Grains with one atom were not considered in the calculations of $\langle s_c \rangle$. We observe a significant evolution of the grain size at lower temperatures. With an increase in the number of deposited

maximum value of the mean grain size is shifted to larger thickness with an increase in temperatures.

The size of the largest grain (s_m), the number of multi-atom grains (n_c), and the number of atoms not connected to any grains (n_i) were also calculated. Figure 3 shows the dependence of the average grain size, the size of the largest grain, the number of multi-atom grains, and the number of isolated atoms on deposited atom number for deposition rate of 1/60 atom per time steps at $T^* = 0.3$ (a) and $T^* = 1.0$ (b). Compare figures 3a and 3b one can find a different evolution of the grain structure during the film growth. In both cases, the number of grains (n_c) increases with an increase the number of deposited atoms. In the initial stage, one observes a local maximum of grain number (n_c) associated with the start of the process of joining of grains. This explanation is supported by an increase in the size of the largest grain (s_m). The coalescence of grains leads to a decrease in the number of grains (n_c) and an increase in the size of largest grain (s_m). Due to the creation of new grain boundaries one observes an additional increase in the number of grains (n_c). As seen from figure 2 the occurrence of the coalescence process is shifted to larger coverage with an increase in temperatures, as a result of higher mobility of atoms.

The number of individual atoms (n_c) also increases with increasing number of deposited atoms in both cases. Figure 4 shows the dependence of the fraction of individual atoms in the inner layers (n_{in}) on the number of deposited atoms for rate 1/60 at different temperatures. With an increase in the coverage, the value of the fraction of individual atoms inside a layer also increases in all cases, but an increase in temperature leads to a reduction in n_{in} . This observation means that at low temperatures there are more individual atoms in the space between the

Examples of this effect are shown by the configurations obtained for different temperatures presented in Figure 5. This difference explains the changes in the biaxial stress in compressive direction (see fig. 1) Individual atoms in a grain boundary generate compressive stress in the films. The larger the number of isolated atoms, the greater is the compressive contribution to the mean biaxial stress. This conclusion is in agreement with the model proposed by Chason et al. [6]. According to the assumption of this model, during continuous growth of films excess atoms are incorporated in the grain boundaries, thereby creating compressive stress in the film. The main reason for the migration of atoms to grain boundary is an increase in the chemical potential of surface of the film in presence of the growth flux.

4 Conclusions

The results of molecular dynamics simulation of the evolution of stress and grains during the deposition of atoms in thin films have been presented. We obtain a characteristic shape of stress curves typical for materials with a Volmer-Weber mode which one obtained from the change in the position of atoms resulting from interatomic potential.

In this paper we founded that:

- The stress has a maximum value when the film become continuous.
- The increase in temperature causes the decrease of the biaxial stress and the mean grain size.
- At a high temperature the majority of individual atoms is on the film surface, but at lower temperature there are in the space between grains. An individual atoms on the grain boundaries generates compressive stresses in films.

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3 Fig.1:

4 Evolution of biaxial stress and normal stress in the growing film as a functioning
5 number of deposited atoms at selected temperatures. Deposition rate is 1/60 atom
6 per time steps.
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11 Fig.2:

12 Dependence of average grain size on the number of deposited atoms at selected
13 temperatures. Deposition rate 1/60 atom per time steps.
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18 Fig. 3:

19 Dependences average grain size, size of the largest grain, number of multi-atom
20 grains, and number of isolated atoms on the number of deposited atom at different
21 temperatures T^* : (a) 0.3 and (b) 1.0. Deposition rate 1/60 atom per time steps.
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27 Fig. 4:

28 Fraction of individual atoms in the inner layers (n_{in}) as a function of the number
29 of deposited atoms at selected temperatures. Examples of deposition rate equals
30 1/60 atom per time steps.
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36 Fig. 5:

37 Examples of three-dimensional configurations obtained from simulation at T^* : (a)
38 0.1 and (b) 0.7. Deposition rate equals 1/60 atom per time steps.
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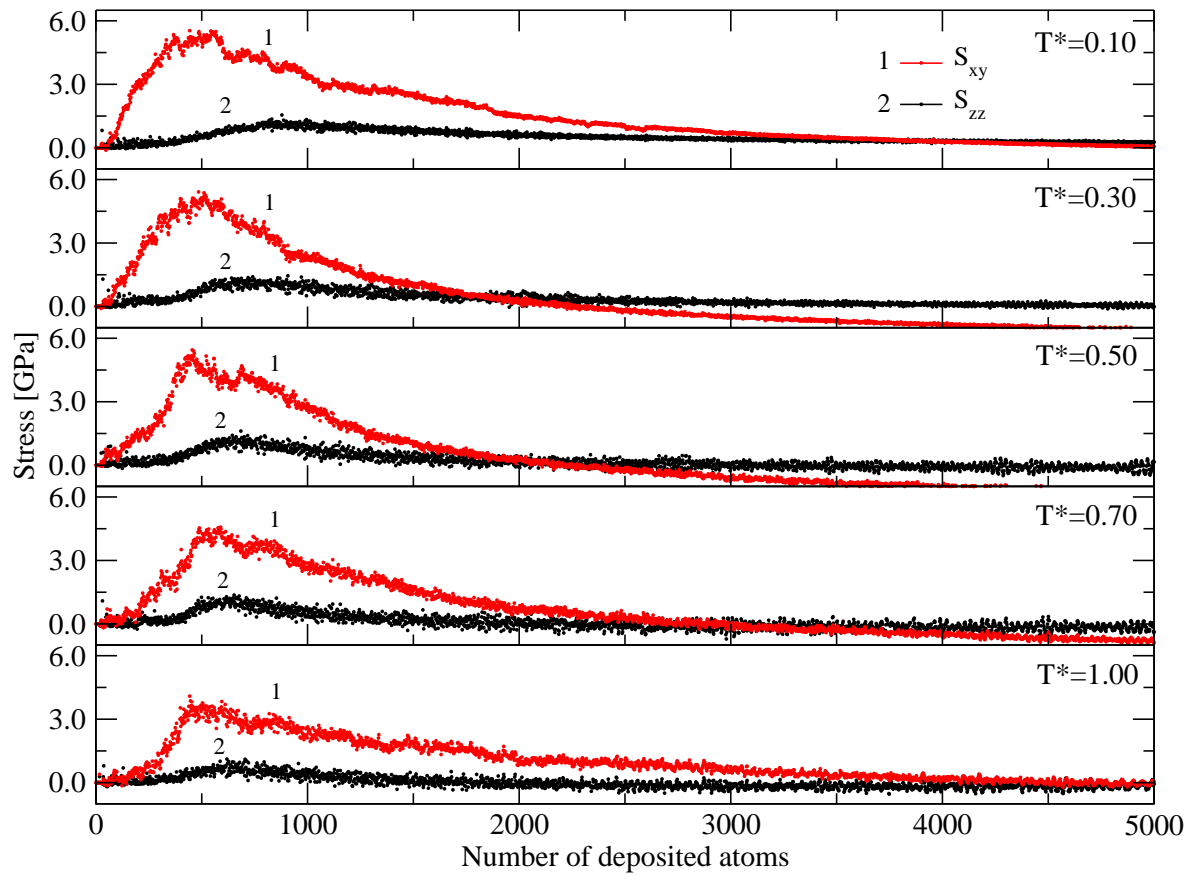


Fig. 1. D. Chocyk ...

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Deposition rate: 1 atom per 60 time steps

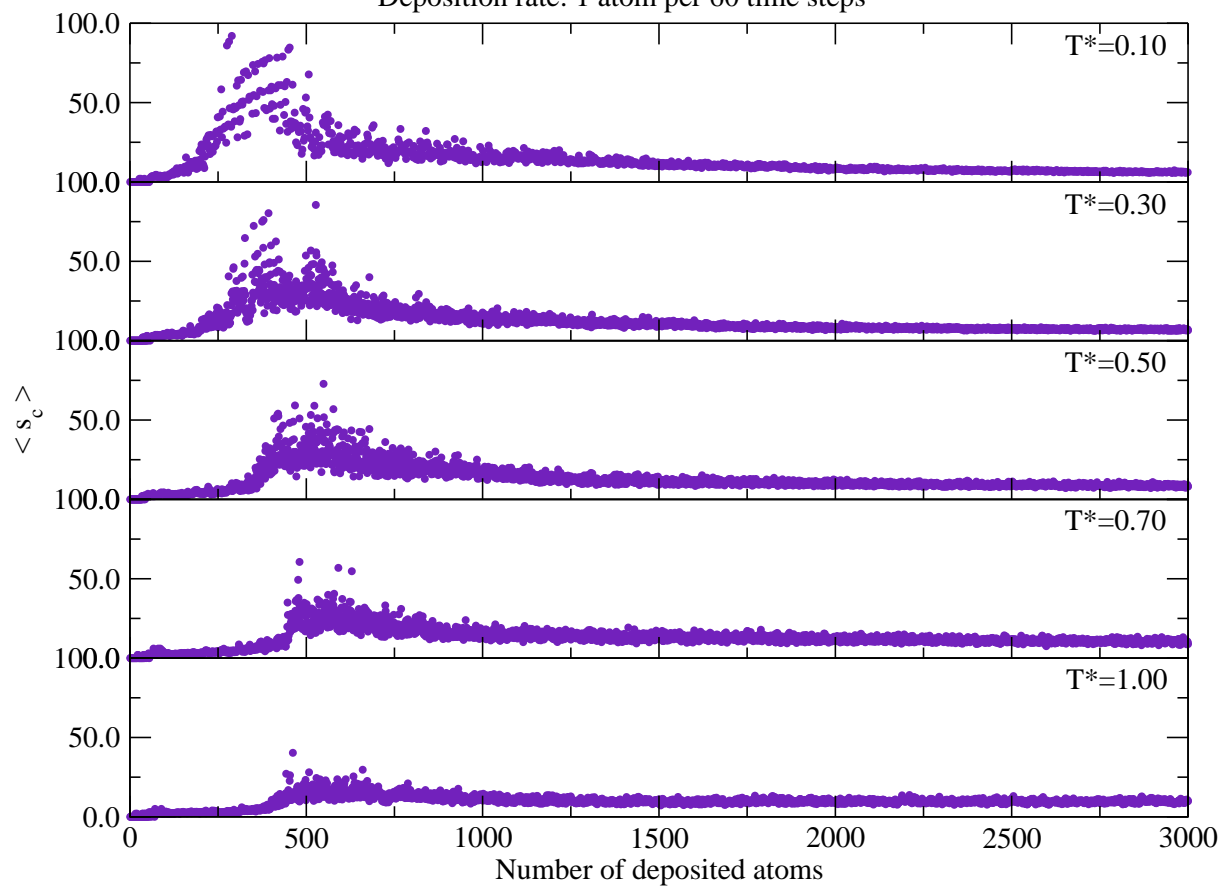


Fig. 2. D. Chocyk ...

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$T^*=0.30$

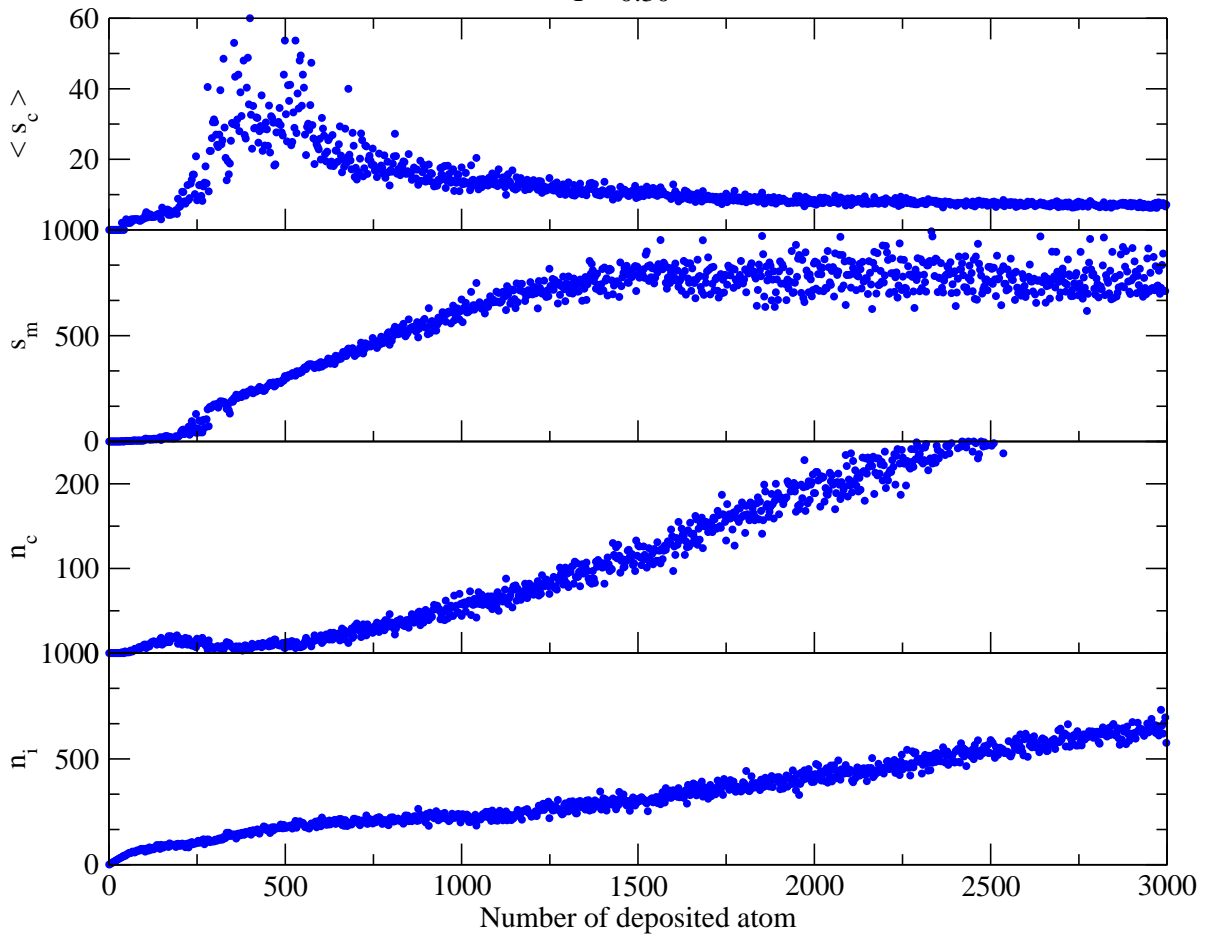


Fig. 3a. D. Chocyk ...

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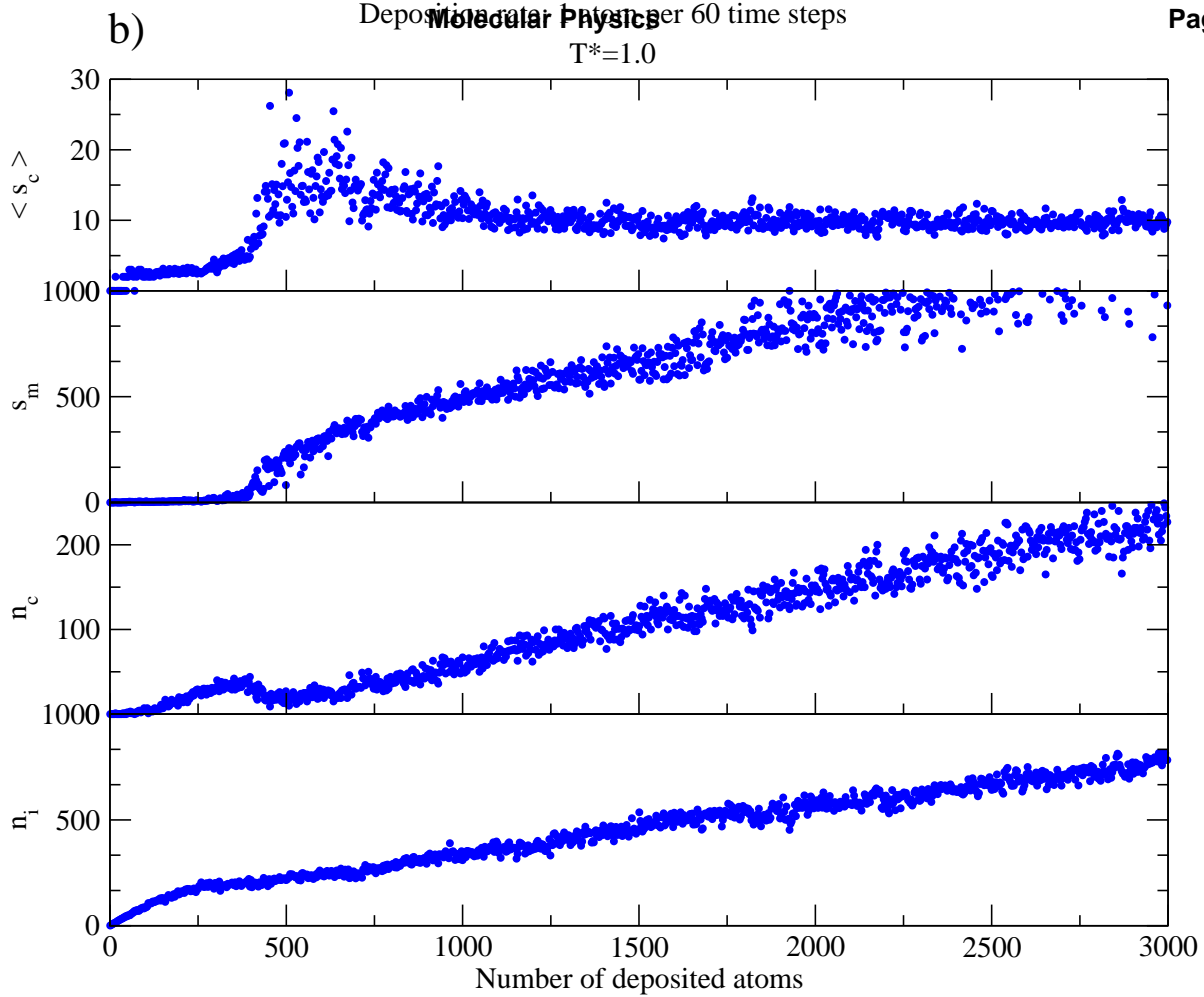


Fig. 3b. D. Chocyk ...

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Deposition rate: 1 atom per 60 time steps

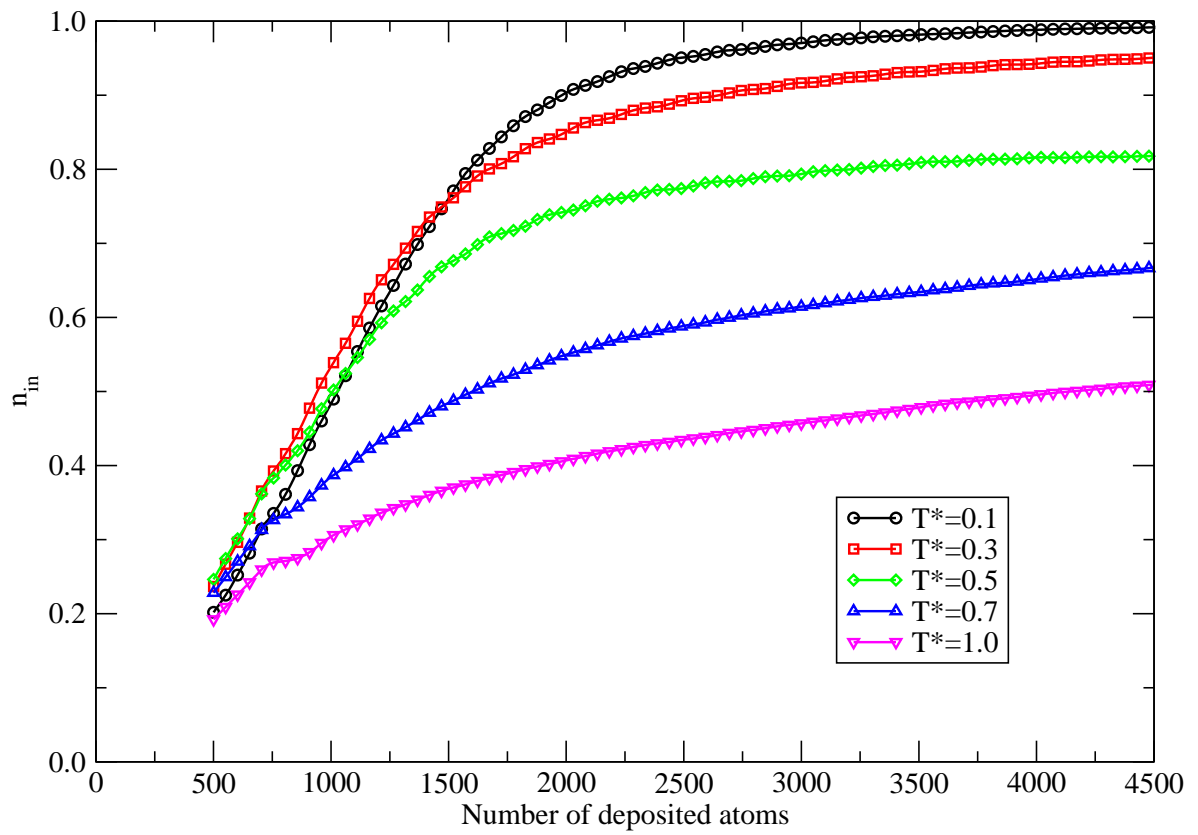
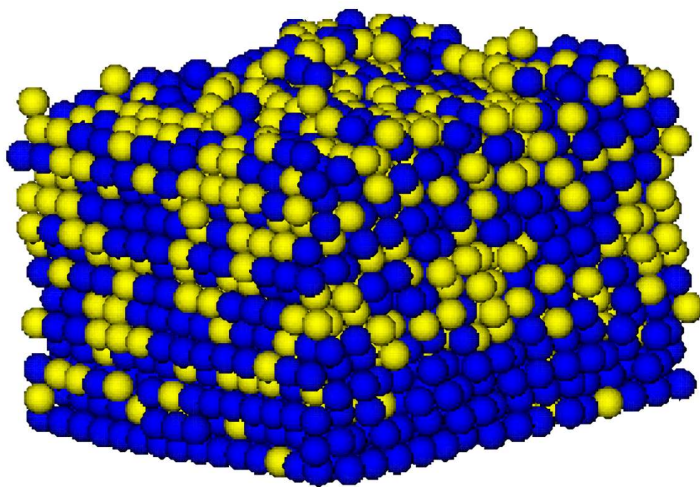


Fig. 4. D. Chocyk ...

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b)

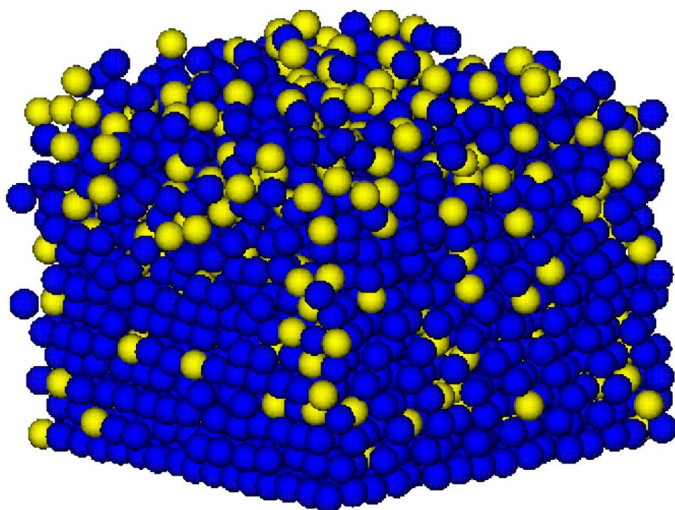


Fig. 5. D. Chocyk ...

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