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Study of initial stages of thin film growth by means of atomistic computer simulation and image analysis: Comparison with experimental data

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ABSTRACT

The initial stages of metal film growth were studied by computer experiment. The structures were generated by an atomistic model combining the molecular dynamics and Monte Carlo approaches and the analysis of obtained structures was performed by image processing algorithms. Film morphology was tested by several methods and it was found that especially Voronoi tessellation and Quadrat Counts methods are best suited for a description of spatial distribution of objects. The same morphological analysis was applied to experimental data – to micrographs of discontinuous films of Ag, Au and In grown on dielectric or graphite substrates. It was found that for small evaporation rates the atomistic model is in a good agreement with experiments. However, when increasing the evaporation rates the effect of secondary nucleation starts to be pronounced and a discrepancy between modelled and experimentally derived morphological results occurs. This effect was demonstrated for indium films with intensive secondary nucleation.

1. Introduction

Discontinuous metal films grown on dielectric substrates [1–3] or composite films of metal/dielectric structure [4,5] are typical examples of systems consisting of a large number of individual objects. These systems can be investigated experimentally, theoretically, or by computer modelling; however the best research results are derived by the combination of several approaches, especially by particle computer simulation based on own experimental data [6–8]. The simulation techniques used are either the molecular dynamics [3,9–11] or the kinetic Monte Carlo approaches [8,12–19]. A large number of questions starting from the nucleation process [20,21], through the morphology of growing discontinuous or semicontinuous films [2,22], to the influence of various material and technological parameters during film growth [7,23], as well as properties of final film structures, was studied experimentally. The reason is that the final micro- and nanostructures are influenced by the growth processes and the ultimate film properties are determined by them. Electrical and optical properties of thin metal (Au, Ag, Cu, Pt, etc.) or semiconductors films are often studied both experimentally and theoretically/computationally [22,24]. There is a close connection between experimental studies and theoretical or computational approaches. Theoretical approaches including computer simulations are necessary to better understand or guide experiments and may allow tuning synthesis parameters in order to achieve better control of film growth. Experimental results, in turn, may be used as inputs to design theoretical models which can be routinely used to plan experiments, thus also allowing for saving time and costs.

The photographs of studied systems from the transmission electron microscopes as well as from AFM or STM microscopes are one of important sources of information about processes in these systems (see e.g. Refs. [6,8]). Various information can be obtained during image analysis of the film micrographs: the characterisation of whole images as mean coverage or concentration of objects, the forms and size distributions of individual objects and the distribution of objects on a substrate (in the case of discontinuous films) or the spatial distribution of objects (in composites).

The aim of the paper is to study the initial stages of metal film

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growth by computer experiment. In our previous papers [18,25] we described an atomistic model combining the molecular dynamics and kinetic Monte Carlo approaches. The present study is devoted to the common analysis of computational and experimental data, the analysis being based on techniques of image processing, with the task to discuss the applicability of both the used models and standard morphological techniques in future studies.

2. Computer experiment

In the contribution a computer experiment for the study of early stages of metal film growth on dielectric substrates is described. The complete computer experiment consists of the following parts:

(i) Preparation of model structures with known properties.
(ii) Image analysis of the simulated structures and critical assessment of results obtained from known information about properties of the model structures.
(iii) Application of the same morphological algorithms on experimental data and the discussion of obtained results.

To prepare the model structures, an atomistic model of initial stages of thin metal film growth was used. The model itself was described in detail in our previous papers [18] and [25]. The model consists of two parts, as the complete time scale for simulation is too large for both the molecular dynamics and Monte Carlo approaches alone. The first part, designed to simulate the nucleation process, is based on the molecular dynamics technique [26]. As the molecular dynamics simulation is extremely slow, it is used for the study of the very beginning of the growth only. The results of the molecular dynamics simulations were used as input for kinetic Monte Carlo simulations of the subsequent stages of the film growth. It means that the initial nuclei or small islands prepared by the molecular dynamics simulations are used as a starting structure for the subsequent Monte Carlo simulations.

In the molecular dynamics part of our atomistic model, various potentials for force calculations between individual atoms were implemented – universal (classic 6–12 Lennard-Jones potential), Stillinger-Weber or Tersoff for semiconductors and Sutton-Chen for metals. Interactions between atoms of different materials were calculated with the help of mixing rules. The temperature of the system was controlled by the Nosé-Hoover thermostat and maintained at a constant value. The computational domain was rather small due to extreme time consumption of the molecular dynamics simulations – it consisted typically of 200×200 to 300×300 atoms with periodic boundary conditions in lateral directions and was 5–7 layers thick, the basic layer being solid. The time steps for the used velocity Verlet mover simulating the adatoms migration were 10^{-15} s; this value was derived from the vibration frequency being of the order of 10^{-13} s. In the model we considered the diffusion and desorption of single atoms, limited diffusion of dimers and we neglected the diffusion of larger aggregates. The diffusion was modelled by thermally activated movement of adatoms to a nearest neighbour position. The simulations were performed for various substrates – mica, amorphous carbon, alumina and Si(111) – and for various metals – mostly Au, Ag, Cu and In. The single atom activation energies of diffusion varied for various materials combinations between 0.18 eV and 0.30 eV while the activation energies of desorption was several times larger.

The forms of nuclei and small objects and the speed of coalescence of larger objects were derived as an output of this part of our atomistic model. These results were used as an input for the subsequent stochastic model.

The following kinetic Monte Carlo part of the model included, besides the results of the molecular dynamics simulations, further physical parameters as the deposition rate, initial energies of metal adatoms, substrate temperature, probability of reflection of impinging atoms, probabilities of migration of individual atoms and small clusters, condensation coefficients of adatoms both on the dielectric substrate and the surface of growing metal film, etc. (details see Ref. [25]). Some of these parameters can be calculated on the basis of corresponding energies obtained in the molecular dynamics part of the model. When studying the laser or plasma pulsed deposition two new parameters, i.e. the pulse length and the repetition rate, must be added, too. The computational domain in this part of atomistic model was larger than in the molecular dynamics simulation – it ranged from 1000×1000 to 5000×5000 pixels with cyclic boundary conditions, which enabled us to derive enough data for the following statistics of both size and spatial distributions of objects. The symmetry of the model substrate corresponds to the given dielectrics. The liquid-like coalescence of objects is considered with the speed tabulated in the molecular dynamics part of the model.

The most important parameters of the whole model are the deposition rate and the substrate temperature [18]. The time scale of the kinetic Monte Carlo modelling is closely connected with the substrate temperature which determines the speed of migration. The deposition rate R represents the intensity of the particle source and is given in arbitrary units as its exact value depends on the combination of other model parameters. Three typical values of the deposition rate – low rate ($R = 1 \times 10^1$), medium rate ($R = 1 \times 10^3$) and very high rate ($R = 1 \times 10^5$) – were used to generate structures and to present their morphological characteristics in the following figures. The generated structures for various mean film thicknesses are shown in Fig. 1. It can be seen from this figure that the concentrations of objects and their spatial distributions are very sensitive to the deposition rate.

3. Results of morphological analysis

The information about the object size distributions can be expressed quantitatively with the help of the distribution of object radii dn/dr. The resulting histograms are presented in Fig. 2. It is clearly seen that a secondary nucleation starts for thicker films when the deposition rate is increased.

For the description of spatial distributions of objects there exist many methods, most of them being based on the theory of mathematical morphology or Fourier optics [27] – e.g. radial distribution function, distribution of nearest neighbours, chord-length distribution, Quadrat Counts method, covariance, Voronoi and Delaunay tessellation, etc. Most of them were tested and results are described in the literature [28–32].

It was found that especially two of these methods – Quadrat Counts method [18,25,33] and Voronoi tessellation [29,30,32] – are very robust and bring the best results. The methods were calibrated by hard-disk and soft-disk models which generate structures ranging from random to completely arranged and in this way the calibration curves of both methods were obtained. The same calibration was performed in the case of composite structures when these methods were applied to sections of composites.

The Voronoi tessellation technique was used as a basic morphological method in the present study and the obtained conclusions were controlled by the other robust morphological characteristic, the Quadrat Counts technique. Analysed image area is decomposed into cells surrounding individual objects in the Voronoi tessellation method. All pixels of each cell are closest to the given object. Large number of so-called features can be constructed on the basis of Voronoi decomposition. Every feature is a scalar morphological characteristic which brings information about the objects distribution in the simplest way. We applied the most