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Modelling of Diffusion-Controlled Pattern Formation in Thin Metallic Film Growth on Crystalline Substrates

Vladimir Kuzovkov[1], *Eugene Kotomin*[1, 2], *and Guntars Zvejnieks*[1]

[1]Institute of Solid State Physics, University of Latvia,
Kengaraga str. 8, Riga, LV 1063, Latvia
E-Mail: kuzovkov@latnet.lv
[2] Max Planck Institut für Festkörperforschung, Stuttgart, Deutschland

1 Introduction

Diffusion-controlled growth of metal nanostructures and thin films on metallic and insulating substrates is important from both theoretical and practical points of view, including many technological applications, such as catalysis and microelectronics. Theory is challenged to describe two modes of thin film growth: monolayer-by-monolayer regime or 3D metal cluster formation [1]. In particular, the Ag thin film growth on MgO substrate is widely studied. Electron microscopy experiments performed at low and moderate temperatures for this system reveal two kinds of *pattern formation*: the spherical compact clusters and worm-like loose cluster growth [2].

2 Modelling of pattern formation

We have performed kinetic MC simulations of the pattern formation as a result of adsorption, diffusion and clustering of mobile metal atoms on a crystalline substrate. Simulations are based on a new complete axiomatic approach (*standard model*) to the kinetics of diffusion-controlled processes with energetic interactions between adsorbates [3]. Pattern formation simulated for submonolayer metal coverage is characterized in terms of the joint correlation functions for spatial distribution of adsorbed atoms and cluster distribution functions. We demonstrate different modes of a monolayer growth, which can be explained and characterized by the ratio of the adatom mutual attraction energy to the substrate temperature. The distinctive average distance between two-dimensional metal islands, estimated by means of a scaling approach, is confirmed by kinetic MC simulations.

In our submonolayer adsorbate modeling, we observe loose adsorbate aggregates for the dimensionless attraction energy $\omega=E_a/k_BT>1.25$, see Fig. 1, and no pattern formation at smaller interaction energies/higher temperatures. Our modeling neglects the role of point defects/impurities, which are often assumed to serve as nucleation sites for aggregates. This is true under following conditions. Let the mean distance between defects be L_d, which is defined by their concentration. If $L_d > L_0$, defects play no role; just adsorbed particles with greater probability meet and join pre-existing aggregates. For high defect concentrations, $L_0 > L_d$, just adsorbed particles meet defects, and form immobile dimers.

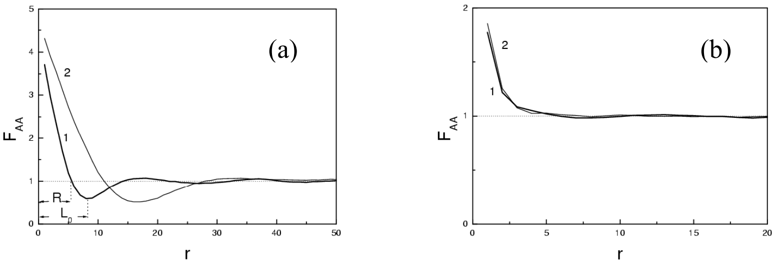

Fig. 1 The joint correlation function vs the relative distance between adsorbate atoms (in units of lattice constant): $C_A=0.1$, hop frequency $d=2^5\ 10^3$ (curve 1) and $d'=2^9\ 10^3$ (curve 2), (a) $\omega=3.0,$ (b) $\omega=1.25$.

For aggregation of other particles such dimers serve very similarly to the result of the encounter of the two mobile adsorbate atoms. As a result, one can expect growth of many small adsorbate aggregates rather than large aggregates. According to our estimates for L_0 presented above and experiments, critical surface defect concentration for Ag/MgO could be of the order of 10^{-4} (in dimensionless units).

3 Conclusion

Our MC modelling demonstrates a considerable difference between simulations assuming the adsorbate concentration to be constant, and for a permanent flux of adsorbates to the surface. The principle point is that adsorbate-adsorbate interactions at relatively low temperatures keep most of particles in their aggregates, which are highly non-equilibrium. This is why no Ostwald-type ripening of small aggregates into larger aggregates takes place at the experimentally observable time scale.

To learn more about adsorbate growth mode, one has to go beyond $\boldsymbol{D}=2$ simulations and model a growth of several metal planes, which is now in progress.

References

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