Interparticle interaction and structure of deposits for competitive model in (2+1)-dimensions

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\textbf{Abstract}

A competitive (2+1)-dimensional model of deposit formation, based on the combination of random sequential absorption deposition (RSAD), ballistic deposition (BD) and random deposition (RD) models, is proposed. This model was named as RSAD\textsubscript{1-s}(RD\textsubscript{f}BD\textsubscript{1-f})\textsubscript{s}. It allows to consider different cases of interparticle interactions from complete repulsion between near-neighbors in the RSAD model ($s = 0$) to sticking interactions in the BD model ($s = 1, f = 0$) or absence of interactions in the RD model ($s = 1, f = 0$). The ideal checkerboard ordered structure was observed for the pure RSAD model ($s = 0$) in the limit of $h \to \infty$. Defects in the ordered structure were observed at small $h$. The density of deposit $p$ versus system size $L$ dependencies were investigated and the scaling parameters and values of $p_\infty = p(L = \infty)$ were determined. Dependencies of $p$ versus parameters of the competitive model $s$ and $f$ were studied. We observed the anomalous behaviour of the deposit density $p_\infty$ with change of the inter-particle repulsion, which goes through minimum on change of the parameter $s$. For pure RSAD model, the concentration of defects decreases with $h$ increase in accordance with the critical law $p \propto h^{-\chi_{RSAD}}$, where $\chi_{RSAD} \approx 0.119 \pm 0.04$.

\textbf{Key words:} Defects, Deposition models: random, random sequential absorption, ballistic, Interparticle interaction, Scaling  
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1 Introduction

Recently the study of the nonequilibrium processes of deposit formation became one of the most active research areas in physics and chemistry because of both fundamental and practical interest [1]. The essential progress in this field was reached owing to application of computer simulations methods for studying the problems of adsorption, formation of thin films and coating. There exists a limited number of computer models, allowing to carry out effective calculations and to establish the main scaling laws for formation of deposits [2].

Among the most popular models for deposit formation simulation are the models of random sequential adsorption (RSA), ballistic deposition (BD) and random deposition (RD) [2,3]. In these models, the particles get fixed after deposition and don’t move, so these models describe the growth processes far from equilibrium. It is also assumed that particles are rigid and can not overlap. In RSA model, particles get fixed at some distance one from another [3,4]. In RSA model with the nearest neighbor (NN) exclusion, configurations with the particles having nearest neighbors are eliminated. In fact, it means existence of some repulsion between particles. In RSA model, all sites in the lattice can be filled with equal probability. However, this requirement is not fulfilled on deposit formation. In this case, the previously deposited particles can screen free sites, located below. So, we named the variant of RSA model for deposit formation as RSA deposition model (RSAD). In BD model, the particles stick at the point of their first contact. It means existence of the short-range attraction. In RD model, the particles deposit without sticking and it means existence of the short-range repulsion. Recently, a number of mixed or competitive models were proposed. They are based on consideration of deposition for different kinds of particles [5,6,7].

In this work, the competitive (2+1)-dimensional model with three kinds of particles, depositing according to the rules of random sequential absorption deposition (RSAD), ballistic deposition (BD) and random deposition (RD), is investigated. This model allows to explore a wide class of systems with different interparticle interactions, which vary from a complete repulsion as for RSAD model to short-range attraction as for BD model.

The paper is organized as follow. The model is described in section 2. In section 3, the scaling behaviour of the deposit density and concentration of the structure defects are discussed for different values of the competitive model parameters. Concluding remarks are presented in section 4.

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2 Model

The competitive (2+1)-dimensional model, which combines models of the random sequential adsorption deposition (RSAD), ballistic deposition (BD) and random deposition (RD), named by as RSAD \(_{1-s} \) (RD\(_f\)BD\(_{1-f}\))\(_s\), where \(s\) and \(f\) parameters are fractions of particles with different kinds of interparticle short-range interaction potentials. A particle of RSAD or BD or RD kind randomly falls straight down onto a growing surface, one at a time, and deposits at a site of the cubic lattice.

Parameter \(s\) characterizes the short-ranged next-near-neighbor repulsion of particles in the deposit, and parameter \(f\) characterizes adhesion of particles. In the extreme case of \(s = 0\) all the particles are of RSAD kind. This case corresponds to the strong repulsion between particles in NN sites and they can deposit only in the next NN sites. When \(s = 1\), \(p = 0\), all the particle are of BD kind and newcomer sticks to the deposits at a point of its first contact. When \(s = 1\), \(p = 1\), all the particle are of RD kind. This case corresponds to existence of the short-ranged repulsion between particles and formation of completely compact deposits.

The density of a deposit may be defined as

\[
p = \frac{N}{V}\tag{1}
\]

where \(N\) is the number of deposited particles, and \(V\) is the volume of deposit.

The value of \(p\) is calculated at a moment when saturation was reached in the system with the volume \(V = L \times L \times L\). At this moment, filling of the system with new particles was terminated.

For RSAD model, the structure of deposit becomes more regular at large deposit height \(h\), and in the extreme case of \(h \to \infty\), the ideal checkerboard-ordered structure get formed. In this ideal structure, the empty lattice sites alternate with occupied ones and each empty (or occupied) site has exactly \(z = 6\) occupied (or empty) NN sites. But at small values of \(h\) there exist some defects in the regular structure (Fig. 1), when NN exclusion number \(z\) is different from 6.

The concentration of defects \(\rho(h)\) may be defined as

\[
\rho(h) = 1 - \bar{\bar{z}}(h)/6, \tag{2}
\]

where \(\bar{\bar{z}}(h)\) is the NN exclusion number averaged over all the occupied and empty sites in the horizontal layer with the constant \(h\).
Fig. 1. Schematic presentation of the deposit for pure RSAD model \( (s = 0) \). Black cubes correspond to the sites filled with particles. In the ideal checkerboard-ordered structure each particle is surrounded by 6 empty sites. The examples of defects are also shown.

The periodic boundary conditions are applied along the horizontal directions \( x \) and \( y \). The size of the base \( L \) was varied within the interval of \( L = 10 - 400 \) and deposit height was \( h \leq 200L \). The values of \( p \) and \( \rho(h) \) were averaged over \( 100 - 1000 \) of different configurations for each fixed set of parameters \( s \) and \( f \).

3 Results and discussion

At the beginning we studied the scaling behavior of the deposit density \( p \) in cubes with varying sizes \( L \times L \times L \). A clear scaling law of the following type was observed:

\[
p = p_\infty + aL^{-1/\nu},
\]  

(3)
Fig. 2. Density of deposit $p$ and scaling exponent $\nu$ versus $s$ in equation 3 at different values of $f$. Insert shows similar plots of scaling amplitude $a_p$ versus $s$.

Here, $p_\infty = p(L \to \infty)$, $\nu$ is the scaling exponent and $a$ is the amplitude.

Figure 2 shows $p_\infty$, $\nu$ and $a$ versus $s$ dependencies at different values of $f$. As we have noted previously, for RSAD model ($s = 0$) in the limits of $L \to \infty$ and $h \to \infty$ the ideal checkerboard ordered structure get formed, that corresponds to $p_\infty = 0.5$. With increasing of $s$ when $f$ is a constant the density of a deposit $p_\infty$ always goes through the minimum at $s = s_{\min}$. The value of $s_{\min}$ increases with decreasing of $f$. The point of $s = 1$ and $f = 0$ corresponds to a pure BD model for which $p_\infty = 0.3000$ (this value in accordance with data [8]). The point of $s = 1$ and $f = 1$ corresponds to the pure RD model, when $p_\infty = 1$ and compact deposit forms.

At first glance, the $p_\infty$ versus $s$ behavior is somewhat anomalous. Really, the effective repulsion between particles decreases with increase of $s$ (at constant $f$). So, we should expect compacting of deposit and increase of $p_\infty$. Another interesting feature is that the scaling exponent $\nu$ decreases abruptly with $s$ decrease in the interval of $s < 0.1$. In fact, we observe two different scaling regimes with large and small values of scaling exponent $\nu$. For the pure RSAD model ($s = 0$), scaling exponent is $\nu_{RSAD} = 8.4 \pm 0.3$. In a regime far from pure RSAD model, the scaling exponent falls within the interval of $\nu = 1.0 - 1.8$. The amplitude $a$ continuously grows with $s$ increase and it approaches zero at $s < 0.1$, which results in large errors of $\nu$ estimation (see insert in Fig. 2).
The nature of unusual behavior of the deposit density $p_\infty$ with variation of $s$ may be understood more clearly from the analysis of defects evolution during the deposit growth. The initial abrupt decreasing of the deposit density with $s$ increase (at $s \lesssim 0.1$) reflects generation of defects in the ideal checkerboard-ordered structure. Naturally this results in loosening of the structure of deposit. The point of density minimum at $s = s_{\text{min}}$ corresponds to a kind of equilibrium between the processes of birth and vanishing of defects. With further $s$ increase, effectiveness of regeneration of the ideal checkerboard-ordered structure decreases. Finally, it results in increase of $p_\infty$ with $s$ increase.

The described behavior of $p_\infty$ can be compared with behavior of the concentration of defects $\rho(z)$. Figure 3 shows the typical map of the defect annihilation heights in the plane of $x - y$ for the pure RSAD model. Here, the lighter color corresponds to the higher deposit coordinate $z$, at which defects disappear in the ideal RSAD structure.

At initial moment of deposit formation, there arise a lot of defects in the ideal RSAD structure. This is a result of the absence of correlations between depo-
Fig. 4. Concentration of RSAD defects \( \rho \) versus the deposit height \( h \) at \( f = 1 \) and different \( s \) and \( L \). The dashed line corresponds to the power equation (4) with the slope \( \chi_{RSAD} \approx 0.119 \pm 0.04 \). The value of \( \rho_\infty \) corresponds to the limit \( h \to \infty \). Insert shows the limiting values of concentration of the RSAD defects \( \rho_\infty \) (at \( h \to \infty \)) and density of a deposit \( p_\infty \) (at \( h = L \to \infty \)) versus \( s \) at different values of \( f \).

sition in the columns at small height of the deposit. These defects can spread in the direction of deposit formation along \( z \) axis and their concentration in each layer \( \rho(z) \) increases or decreases owing to the processes of defect birth (at \( s \neq 0 \)) or annihilation, respectively.

Figure 4 shows the examples of defects concentration \( \rho \) versus deposit height \( h \) dependences for the case when \( f = 1 \). The similar dependences were also observed for other values of \( f \).

For pure RSAD model \((s = 0)\) the concentration of defects \( \rho \) continuously decreases with increase of the deposit height \( h \). In the limit of \( h \to \infty \) the defectless checkerboard-ordered structure get formed. In the latter case, the obvious scaling behavior of \( \rho(h) \) dependencies is observed for systems with different size of base \( L \). We believe that a simple power law can be applied for
description of $\rho(h)$ behavior in the limit of $L \to \infty$

$$\rho \propto h^{-\chi_{RSAD}}$$

where $\chi_{RSAD}$ is the scaling exponent of defects annihilation for the pure RSAD model.

This behaviour is rather similar to the critical annihilation of inter-domain boundaries in the model of competitive growth by Saito and Muller-Krumbhaar (SMK) [9] (see also, [10,11]).

Prediction of the scaling exponent in critical annihilation is not trivial. For normal Brownian motion of annihilating defects the scaling exponent should be $\chi_B = 1/2$ [12]. In 1 + 1 dimensional SMK-model another scaling exponent $\chi_{SMK} \approx 2/3$ [9] was observed. This result was explained by existence of different mechanisms of the critical annihilation of defects in SMK and random walk motion models. It was conjectured that the lateral displacements of inter-domain boundaries in the SMK model are controlled by the same processes as those causing roughening the outer interface width and $\chi_{SMK} \approx \alpha$, where $\alpha$ is the roughening exponent. On the other hand, the value of $\alpha$ can reflect the growth mechanism, geometrical confinement of growing interface, space dimension, etc.[13]. For example, for usual models of KPZ class of universality [14] the roughening exponent is $\alpha = 1/2$ in 1 + 1 dimension and is $\approx 0.2 - 0.4$ in 2 + 1 dimension [2].

For pure RSAD model ($s = 0$) and an isotropic system ($L \times L \times L$), we can estimate the value of the scaling exponent in Eq. 4 as $\chi_{RSAD} = 1/\nu_{RSAD} \approx 0.119 \pm 0.04$, where $\nu_{RSAD} = 8.4 \pm 0.3$ is the scaling exponent for the density of deposit in Eq. 3. Indeed, we believe that the scaling changes in the deposit density $p$ and in the concentration of defects $\rho$ are controlled by the same process, and $p(L) \propto \rho(L)$. The dashed line in Fig. 4 corresponds to the slope, estimated from scaling of the deposit density. But for anisotropic systems $L \times L \times h$ ($h > L$), the concentration of defects decrease more quickly than it is predicted by the power law in Eq. 4, and the most obvious deviations are observed with increase of the system anisotropy degree at high $h$ (see Fig.4).

In a general case of the competitive model RSAD $1-s$ (RD$_f$BD$_{1-f}$)$_s$, the patterns of defect distributions in a deposit can be more complicated. Here, defects can birth or can vanish and competition between these processes results in formation of a certain defected structure with the finite concentration of defects. For mixed model at $s \neq 0$, the critical behavior of (4) type disappears. In the limit of indefinitely large $h$ ($h \to \infty$), the concentration of defects remain finite and has a nonzero value $\rho_\infty$. With $s$ increase, which corresponds to the weakening of repulsion between the particles, increase of $\rho_\infty$ is observed (See insert in Fig.4), and in the limit of pure RD model, ($s = 1, f = 1$), $\rho_\infty = 1,$
because deposit is compact in RD model and even local checkerboard-ordered structure is absent.

4 Conclusions

It was demonstrated that implementation of the RSA rules for 3d- growth of deposit on a cubic lattice with the nearest-neighbor exclusions results in formation of a checkerboard-ordered structure. The initial disorder of this structure critically disappears when the deposit grows and, finally, an ideal checkerboard-ordered structure gets formed. In a competitive RSAD$_{1-s}(\text{RD}_f\text{BD}_{1-f})_s$ model, the deposit density goes through the minimum with variation of $s$ parameter at constant $f$. Such behavior reflects the influence of competitive processes related with the birth and annihilation of defects.

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