Percolation in models of thin film depositions

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We have studied the percolation behavior of deposits for different (2+1)-dimensional models of surface layer formation. The mixed model of deposition was used, where particles were deposited selectively according to the random (RD) and ballistic (BD) deposition rules. In the mixed one-component models with deposition of only conducting particles, the mean height of the percolation layer (measured in monolayers) grows continuously from 0.898 32 for the pure RD model to 2.605 for the pure BD model, but the percolation transition belongs to the same universality class, as in the two-dimensional (2D) random percolation problem. In two-component models with deposition of conducting and isolating particles, the percolation layer height approaches infinity as concentration of the isolating particles becomes higher than some critical value. The crossover transition from 2D to 3D percolation was observed with increase of the percolation layer height.

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The thin-film formation processes by deposition of particles on a substrate are of great interest both from theoretical as well as experimental point of view [1,2]. The different aspects of this problem are important in the technical applications for production of thin-film devices, metal-insulator mixture films, composite films with specific physical properties, etc. [3].

The rather important field of investigation is related to the electrical conductivity of thin films, which depends strongly on their morphology and microstructure. Many works were devoted to investigations of the fractal, percolation and electrical properties of thin films and deposits [4]. It was shown that the percolation transition in very thin (quasi-two-dimensional) films belongs to the same universality class as in the random percolation problem [5,6]. The film electrical conductivity shows also a clear transition from the two-dimensional to three-dimensional behavior with film thickness increase [7,8]. Some correlations were observed between the conductivity and porosity for deposits grown in a model of ballistic deposition [9]. Jensen *et al.* [10] and Family [11] investigated in their numerical simulation works the percolation behavior for different models of submononolayer deposits on two-dimensional substrates.

The purpose of this work is to study the percolation behavior for different lattice models of three-dimensional deposits growing on the plane substrates. The spanning cluster forms in the substrate plane. The percolation in deposits has a correlated character, because the sites of lattice get filled dynamically during the growth in accordance with the deposition rules. In our model the particles are modeled by unit cubes. They are deposited on an initially flat horizontal surface on the x-y plane of size $L\times L$. The particles come down vertically along the $-z$ direction with the integer x,y coordinates and are deposited on the substrate either by the ballistic deposition (BD) process or by the random deposition (RD) process or by a mixture of both the processes. In the RD process a particle comes down vertically till it lands over a particle on the substrate where as in the BD process the particle gets stuck to the substrate when any of its four vertical sides comes in contact with any previously deposited particle of the substrate or it directly lands on the substrate. In a mixed RD+BD model, the s fraction of particles is deposited according to the BD rule and the remaining $1-s$ fraction is deposited according to the RD rules. We call this model as: BD_sRD_{1-s} model. The parameter s allows a continuous tuning between RD model with no interaction between particles and BD model with strong short-ranged interaction between particles. The model of this type or other similar models where different kind of interactions between particles may exist are widely used for simulation of structure of thin films with realistic morphology [12].

In the one-component model all particles are considered to be conducting. In the two-component model we have a f fraction of insulating particles and the $1-f$ fraction of conducting particles.

Particles are deposited on the substrate one after another and the average height of the deposit grows. Conduction takes place between two conducting particles when they have one surface in contact. We stop the growth process when the deposit starts conducting for the first time in the direction parallel to the surface. At this percolation point, a spanning cluster across the system is formed along the x or y direction (Fig. 1). The percolation point is easily checked by a Hoshen-Kopelman algorithm [13].

During the deposition process, the time elapsed is measured in units of the number of equivalent complete layers deposited. Therefore, N particles have been deposited in time $t=N/L^2$. On the other hand the mean height of the deposit at time t is $\overline{h}=\Sigma_{x,y}h_{xy}(t)/L^2$. The percolation density is the volume fraction p of the conducting particles at the percolation point i.e., the ratio of the number of conducting particles N_c and the total volume of the deposit $p=N_c/(\overline{h}L^2)$. In one-component model $N_c=N$ and $p=t/\overline{h}$. For RD model the bulk of the deposit is compact (without any pores in the vertical columns) but it has a rough interface. Therefore the

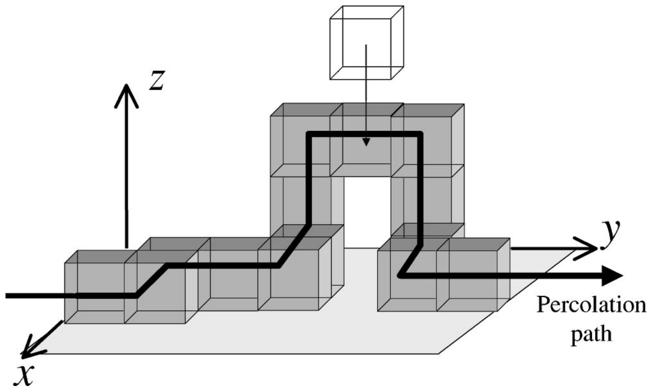

FIG. 1. Scheme of percolation cluster formation for (2+1)-dimensional deposition model.

RD limit at $s=0$ corresponds to $p=1$. For BD model at $s=1$ the deposit is porous and consequently $p<1$. For a bicomponent model we have a mixture of conducting and insulating particles. Here the total density of the deposit including conducting and insulating particles is $p_{total}=N/(\overline{h}L^2)$ and $N_c=N(1-f)$ and $p=t(1-f)/\overline{h}=p_{total}(1-f)$.

For finite extensions (L) of the substrates the percolation height $\overline{h}(L)$ and $p(L)$ are L dependent. The values of $\overline{h}(L)$ and $p(L)$ are determined for different substrate sizes L varied from 8 to 2048 and the periodical boundary conditions were applied in deposition rules along the directions x and y. Results were averaged over 100–5000 different runs, depending on the size of the lattice and required precision.

In analogy with the corresponding finite size behaviors in the ordinary percolation, we assume the following relations:

$$p(L)=p_\infty+a_pL^{-1/\nu_p} \tag{1}$$

and

$$\overline{h}(L)=\overline{h}(\infty)+a_hL^{-1/\nu_h}, \tag{2}$$

where $\nu_p,\nu_h\approx4/3$.

The probability that a particle is deposited along a particular vertical line on the x-y plane is $1/L^2$ which is very small when L is large such that the mean height $\overline{h}$ is maintained at a fixed value when N particles are deposited. This implies that in the RD model the number h of particles in an arbitrary column of particles follow a Poisson distribution

$$P(h)=(e^{-\overline{h}})(\overline{h}^h)/h!. \tag{3}$$

Therefore, the probability of an empty column $(h=0)$ is equal to $P(0)=e^{-\overline{h}}$. In percolation point

$$P(0)=1-p_{2d}=e^{-\overline{h}}, \tag{4}$$

where p_{2d} is the percolation threshold for the square lattice site percolation problem.

Taking into account the finite size scaling behavior of p,

$$p_{2d}(L)=p_{2d,\infty}+a_{2d,p}L^{-1/\nu_p}, \tag{5}$$

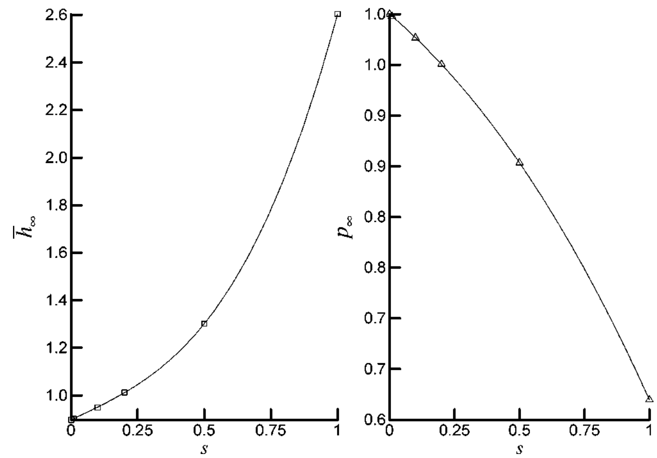

FIG. 2. Plots of height $\overline{h}_\infty$ (lattice units) and density p_∞ of deposit in percolation point vs parameter s for one-component mixed BD_sRD_{1-s} model. The data error is of order of data symbol size. The lines serve as a guide to the eye.

where $p_{2d,\infty}=0.592\,746\ldots$ is the percolation concentration in the limit of infinite system $(L\to\infty)$ and $\nu_p=4/3$ is a correlation length scaling exponent [14], we obtain

$$\overline{h}(L)=-\ln(1-p_{2d,\infty}-a_{2d,p}L^{-1/\nu_p}) \tag{6}$$

$$=\overline{h}_\infty+\ln\left(1-\frac{a_{2d,p}}{(1-p_{2d,\infty})L^{1/\nu_p}}\right)\approx\overline{h}_\infty-a_hL^{-1/\nu_h}, \tag{7}$$

where $\overline{h}_\infty=-\ln(1-p_{2d,\infty})\approx0.898\,32$, $a_h=a_{2d,p}/(1-p_{2d,\infty})$, $\nu_h=\nu_p$.

We see that for pure RD model $\nu_h=\nu_p=4/3$. So, the RD model belong to same class of universality as the two-dimensional random percolation model. This fact reflect the small mean height of (2+1)-dimensional random deposit $\overline{h}_\infty\approx0.898\,32$, which is only slightly higher than mean height of two-dimensional random deposit $\overline{h}_\infty\approx0.592\,746$.

On the basis of numerical simulations we estimate that for pure BD at $s=1$, $\overline{h}_\infty=2.605\pm0.005$ and $p_\infty=0.620\pm0.005$ in the limit $L=\infty$. Using these asymptotic values we plot $\overline{h}_\infty-\overline{h}(L)$ and $p_\infty-p(L)$ vs L on double logarithmic scales and in both cases plots correspond to the slope of $-3/4$, which means $\nu_h=\nu_p\simeq4/3$.

Percolation height $\overline{h}_\infty$ and the percolation densities p_∞ are similarly calculated for the mixed BD_sRD_{1-s} model varying the mixing parameter s and plotted in Fig. 2. The height of the deposit $\overline{h}_\infty$ increases and its density decreases smoothly with increase of the fraction of deposited BD particles. In the limit of pure RD model, the theoretical value $\overline{h}_\infty\approx0.898\,32$ is observed well. Same as both pure BD and RD models, the mixed BD_sRD_{1-s} also displays the scaling behavior described by Eqs. (1) and (2) with scaling exponent $\nu_h=\nu_p\approx4/3$. Using the calculated $\overline{h}_\infty(s)$ and $p_\infty(s)$ dependencies and substituting $\nu_h=\nu_p=4/3$ into Eqs. (1) and (2) the coefficients a_h and a_p versus s were obtained. Both of these coefficients a_h and a_p increase with s. It is important to note

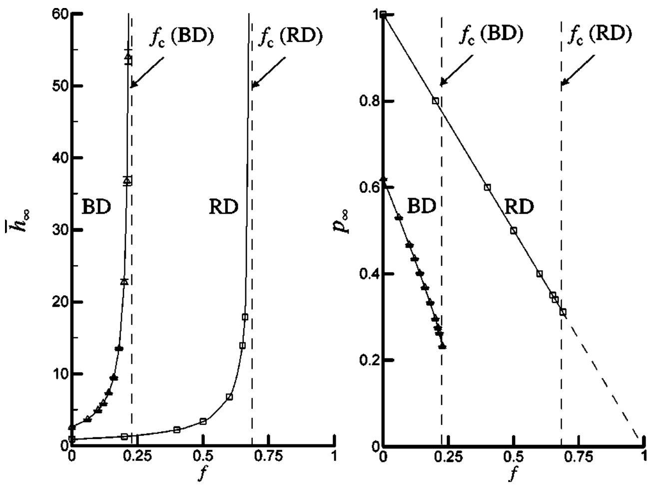

FIG. 3. Plots of height $\overline{h}_\infty$ (lattice units) and density p_∞ of deposit in percolation point vs fraction of isolating particles for two-component BD and RD models. In cases when it is not show directly the data error is of order of data symbol size. The solid lines serve as a guide to the eye. Vertical dashed lines show the critical concentrations which are $f_c = 0.227 \pm 0.001$ and $f_c = 0.70 \pm 0.01$ for BD and RD models, respectively.

that for the pure RD model $a_p = 0$. It means that there is no finite size scaling for the RD model, as for any L at $p = 1$ the deposit is compact without pores by definition.

We can conclude that the mixed $\mathrm{BD}_s\mathrm{RD}_{1-s}$ model, presumably, belong to same class of universality as the two-dimensional random percolation model at any value of s.

Figure 3 presents the deposit height $\overline{h}_\infty$ and density of conducting particles p_∞ in the percolation point estimated in the limit of $L \to \infty$ vs fraction of isolating particles f. The dependencies of $\overline{h}_\infty(f)$ show the typical percolation behavior: as f reaches some critical value f_c the value of $\overline{h}_\infty$ goes to infinity; it means that there is no percolation at any finite height of deposit. The estimated values of critical concentrations of the isolating particles are $f_c(\mathrm{BD}) = 0.227 \pm 0.001$ for the BD model and $f_c(\mathrm{RD}) = 0.70 \pm 0.01$ for RD model.

For the RD model, the total density of particles is $p_{total} = 1$ by definition, and, so, the density of conducting particles is $p = p_{total}(1-f) = 1-f$. The linear law of p_∞ decrease with f increase is actually observed in simulation data for the RD model, this law is rather close to linear for the BD model (Fig. 3). In the critical point $f = f_c$, the density of conducting particles is equal to $p_{c,\infty} = 0.232 \pm 0.001$ for the BD model, and $p_{c,\infty} = 0.30 \pm 0.01$ for the RD model. This value for the RD model is very close to percolation concentration for the random percolation on a simple cubic lattice $p = 0.311\,609$ [15]. The estimated value of the total density of deposit $p_{total,\infty} = 0.300 \pm 0.001$ for the BD model coincides with the previously reported data for the deposit density extrapolated to the infinite-system limit for the BD model [16].

The scaling exponents ν_h, ν_p obtained by the least square fit of Eqs. (1) and (2) versus f are presented at Fig. 4 for the BD and RD models. All fitting procedures were done at the fixed values of p_∞, two free parameters and correlation coefficients were higher than 0.998.

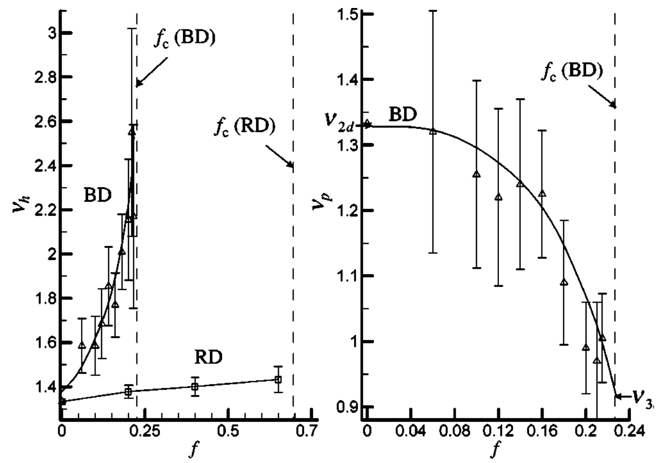

FIG. 4. Plots of scaling exponents ν_h and ν_p in Eqs. (1) and (2) vs fraction of isolating particles for two-component BD and RD models. The solid lines serve as a guide to the eye. Vertical dashed lines show the critical concentration which are $f_c = 0.227 \pm 0.001$ for BD model and $f_c = 0.70 \pm 0.01$ for BD model.

For the RD model, the value of p is independent from system size L by definition, so, only ν_h dependency is presented at Fig. 4. The exponent ν_h for the BD model continuously grows with s from $\nu_h \approx 4/3$ characteristic for single-component model to $\nu_h \approx 2.4$ near critical point $f = f_c = 0.227$.

The correlation length exponent ν_p for BD model decreases from $\nu_p = \nu_{2d} = 4/3$ characteristic for the 2D systems (at $f = 0$) to $\nu_p = \nu_{3d} = 9/10$ characteristic for the 3D systems (at $f = f_c = 0.227$) [14]. This behavior can be easily understood, as far as the height of percolation deposit at high-f increases and approaches the infinity at the critical concentration of isolating particles. So, the BD-model percolation of deposit in the vicinity of $f = f_c = 0.227$ may be considered as a percolation in a three-dimensional system and the universality class of this percolation model is presumably the same as for the random percolation model.

The dependence of ν_p vs f for the RD model is not very pronounced at $f < 0.15$ and the precision of ν_p determination at higher f is rather low, as the respective amplitude a_p continuously grows with f. We can suppose the existence of 2D-to-3D percolation crossover for the BD model in the vicinity of $f = f_c = 0.227$. Note, that the existence of the 2D-to-3D percolation crossover was experimentally supported in random metal-insulator mixture films when thickness of the films deposited was increased [8].

As far as at $f < f_c$, $(L \to \infty)$ the height of a percolation cluster $\overline{h}_\infty$ remains finite even for infinitely large substrate dimension, it is interesting to check p_∞ vs $\overline{h}_\infty$ for existence of scaling:

$$p_\infty = p_{c,\infty} + a_{ph}\overline{h}_\infty^{-1/\nu_{ph}}. \tag{8}$$

Figure 5 shows the log-log presentation of $p_\infty - p_{c,\infty}$ vs $\overline{h}_\infty$ for the BD and RD models. For the BD model, we put $p_{c,\infty} = 0.232$. The solid line 1 corresponds to best fit of Eq.

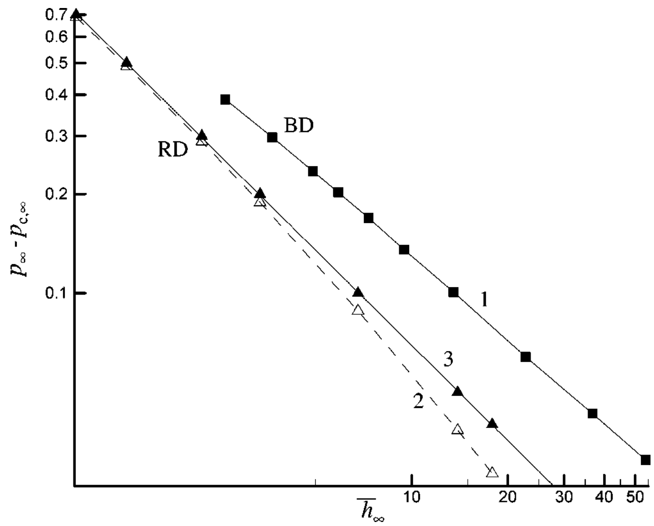

FIG. 5. Log-log plot of $p_\infty - p_{c,\infty}$ vs $\overline{h}_\infty$ (lattice units) for two-component BD (squares) and RD (triangles) models. See the text for the details.

(8) to the data (filled squares) with parameters $\nu_{ph}=1.20\pm0.01$ and $a_{ph}=0.883\pm0.001$.

Putting the value $p_{c,\infty}=0.311\,609$ for the RD model, which is equal exactly to the concentration for the random percolation problem on simple cubic lattice, we obtain data represented by the open triangles. The dashed line 2 is drawn as a guide to the eyes and the scaling is rather poor. But when we put a somewhat higher value $p_{c,\infty}=0.30$ (remind that the value obtained from $\overline{h}_\infty$ vs f dependencies is $p_{c,\infty}=0.30\pm0.01$) we get the data represented by filled triangles. Now the scaling is rather good and the solid line 3 corresponds to best fit of Eq. (8) to the data (filled triangles) with parameters $\nu_{ph}=1.04\pm0.01$ and $a_{ph}=0.633\pm0.004$.

In summary, we have investigated the percolation in the direction parallel to the surface for different models of deposition layer formation. In one-component models with deposition of only conducting particles, the height of deposits in the point of percolation is finite and is $\overline{h}_\infty=0.898\,32$ for the RD model $\overline{h}_\infty=2.605$ for the BD model. The mixed BD_sRD_{1-s} model, presumably, belongs to the universality class of 2D-random percolation problem. In two-component models with conducting and isolating particles, the percolation layer height $\overline{h}_\infty$ can be varied in wide range by tuning of a concentration of the isolating particles f and a crossover transition from 2D to 3D percolation is observed with increase of $\overline{h}_\infty$. The percolation in layer is impossible when f exceeds some critical concentration f_c. The weakening of interparticle interaction results in increasing of threshold value of f_c, from 0.227 for BD model ≈ 0.7 for RD model.

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