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Surface diffusion and interaction of adsorbed barium atoms on the (011) face of molybdenum

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The surface diffusion of barium adsorbed on the (011) face of molybdenum has been investigated in the region of submonolayer coverage. Plateau segments are observed in the diffusion distributions at adsorbed-atom concentrations at which ordered two-dimensional lattices of the type $c(6 \times 2)$ and $c(2 \times 2)$ are formed. The diffusion coefficient and diffusion activation energy change nonmonotonically with change of the degree of coverage of the surface by barium and have extremal values at the indicated concentrations. It is observed that in diffusion from a layer thicker than a monolayer, there is propagated along the surface not a dense monolayer (the unrolling-carpet mechanism) but a submonolayer phase $c(2 \times 2)$. The results are discussed on the basis of the data obtained previously on the features of interaction of adsorbed atoms (adatoms) and on phase transitions in submonolayer films of barium on Mo(011).

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INTRODUCTION

Studies of the atomic structure of adsorbed films, the kinetics of adsorption and desorption, and the influence of the films on the work function of the surface reveal an extremely complicated nature of the interaction of the particles adsorbed on the surface.¹

A number of studies²⁻⁹ show that study of surface diffusion on single-crystal substrates can also be an important source of information on the interaction of adsorbed atoms (adatoms). Up to the present time very few such studies have been carried out. This is due to difficulties in setting up the experiments, in which it is necessary to measure with high accuracy the distribution of the diffusing material on a surface which has a known atomic structure and a negligible number of defects. Such experiments are of course necessary also in order to shed light on the mechanisms of the diffusion processes themselves.

In the present work we have investigated the diffusion of barium on the (011) face of molybdenum. This system was chosen because detailed information has been

obtained previously by various methods regarding the properties of barium films on this substrate (the atomic structure of the films and the phase transitions in them, and the concentration-dependence of the work function and heat of adsorption).³ Preliminary results of the present work have been published elsewhere.⁹

METHOD

The experiments were carried out in glass apparatus under conditions of ultrahigh vacuum. The technique used² was based on recording the distribution of the work function φ over the substrate. With the aid of the known dependence $\varphi(n)$, where n is the concentration of adatoms, the transition is accomplished from φ to the concentration distribution. The work function was measured with an accuracy 0.02 eV by a contact-potential-difference method by means of an electron gun (Fig. 1) with a small exit aperture ($\sim 20 \mu\text{m}$). To avoid spreading of the electron beam, the apparatus was placed in a longitudinal magnetic field H .

The system permitted the surface to be probed by the electron beam in two mutually perpendicular directions.

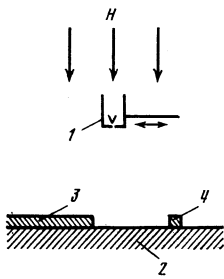


FIG. 1. Diagram of experiment: 1—movable electron gun, 2—substrate, 3—film of adsorbate in the form of a step, 4—in the form of a band.

Along the [001] direction (the x axis) the gun could be displaced by a micrometer screw through a siphon. Here the gun position was determined by means of a microscope. Displacement of the beam in the perpendicular direction (the y axis) was accomplished by rotation of the magnet with respect to the apparatus. The relation between the magnet-rotation angle and the beam displacement was determined in special calibration experiments. The lateral resolution of the technique is $\sim 20 \mu\text{m}$.

The substrate consisted of a single crystal of molybdenum of dimensions $9 \times 2.7 \times 0.07 \text{ mm}$. After grinding and mechanical polishing, the crystal was subjected to electropolishing in a 5% solution of NaOH to remove the damaged layer. The deviation of the surface plane from the (011) face did not exceed $10'$. The crystal was mounted on an arm extending into a Dewar. It could be cooled by liquid nitrogen and heated by passage of alternating current up to temperatures close to the melting point. The crystal temperature was determined by means of a W-Re thermocouple. The time of heating the crystal to the working temperature did not exceed 5 sec. The heating current was stabilized, and the temperature of the crystal was constant in time within $\pm 5^\circ$.

The crystal and the barium evaporator, which has been described previously,³ were carefully outgassed. The experiments were carried out at a pressure of active residual gases 10^{-11} – 10^{-12} Torr. The vacuum was estimated from the change with time of the image of a clean tungsten tip on the screen of a field-emission electron-projection pressure gauge.

The experimental procedure was as follows. By evaporation of barium through a movable mask having a set of openings, the initial distribution of adsorbate was specified in the form of step or a narrow band $\sim 100 \mu\text{m}$ wide (Fig. 1). In some experiments an equilibrium layer of barium (a pedestal) was evaporated onto the surface beforehand, and then a film in the form of step or a band was evaporated on this pedestal. The technique has been described in detail previously.²

The experimental data for determination of the diffusion coefficient D and the diffusion activation energy E_d associated with various concentrations were obtained in the following way (Fig. 2). On a clean substrate or on a pedestal with a given concentration n_p , was evaporated a narrow band of barium, the concentration Δn in which was $1/15$ of a monolayer, and in some experiments even less. Then the time dependences of the concentration at the maximum of the distribution were recorded for various temperatures (a range of temperatures 375–

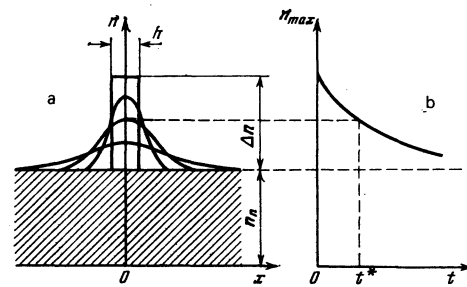


FIG. 2. Illustration of the method of determining D and E_d . a—The dependence $n(x)$ in diffusion from a band for different heating times t with $T = \text{const}$, b—concentration at the maximum of the distribution as a function of t .

500 K). It was assumed that the diffusion parameters in the narrow coverage interval Δn characterizing the excess of the concentration in the band over that in the pedestal do not depend on n .

The diffusion coefficient was calculated according to the equation¹⁰

$$D = Q_0^2 / 4\pi \left(\frac{\Delta n}{2} \right)^2 t, \quad (1)$$

where $Q_0 = \Delta n h$ (Δn is the initial concentration in the band and h is the width of the band). The activation energy was determined from the Arrhenius equation

$$1/t^* = C \exp(-E_d/kT). \quad (2)$$

In these equations t^* is the time of decrease of the concentration at the maximum of the distribution by a factor of two (i.e., to a value $\Delta n/2$). In constructing the dependences $D(n)$ and $E_d(n)$ the values of D and E_d obtained by this means were assigned to concentrations $n = n_p + \Delta n/2$.

Special experiments were carried out in which the possibilities of loss of barium from the surface under study as the result of evaporation and also by diffusion to the backside or into the volume of the crystal were checked. These experiments showed that for the values of t and T used these effects can be neglected.

In the study of diffusion, the perfection of the substrate is of great importance. Although all possible measures were taken to polish the surface carefully and to assure that it had the minimal departure from the specified direction, this of course does not mean that it was completely free of steps and other defects. As an objective indication of the degree of perfection of the surface we can cite the following facts: 1) on the crystal samples used, a low-energy electron-diffraction pattern with sharp reflections is obtained. It follows from this that the characteristic size of the portions of the surface with perfect structure are in any case no less than the coherence width of the electrons (hundreds of angstroms); 2) in photographs obtained by the method of scanning electron microscopy with a resolution $\sim 300 \text{ \AA}$, no defects were observed on the surface of the samples investigated; 3) the nonuniformity of the surface in work function was less than 0.02 eV.

As Butz and Wagner⁵ have established, steps in the surface can have a substantial influence on the surface-

diffusion process. In our experiments it was observed that the boundary of a film which initially was straight can become bent in the course of diffusion. This apparently is also related to the presence on the surface of steps or other defects. The diffusion studies were carried out in those portions of the crystal where this effect was least marked. It was also established that the surface defects appear significantly more weakly when diffusion occurs on the background of a previously deposited pedestal (even with a pedestal concentration $1/20$ of a monolayer). Apparently in this case there is a distinct healing of the defects (they are filled by adatoms).

Analysis of the results permits the conclusion that in our experiments the surface defects, although they could affect to some degree quantitatively the diffusion parameters determined, could not qualitatively change the dependences obtained.

EXPERIMENTAL RESULTS

Let us consider first the data referring to the case in which the maximum barium concentration in the initial distribution $n(x)$ does not exceed a monolayer ($n_m = 6 \times 10^{14} \text{ cm}^{-2}$, degree of coverage $\theta = 1$). Figure 3 shows one of the typical distributions $n(x)$ obtained immediately after evaporation of barium at liquid-nitrogen temperature (77 K). If barium is evaporated at room temperature of the substrate, then during the evaporation time (~20 min) the film boundary is displaced by a distance $\approx 400 \mu\text{m}$, which indicates a substantial mobility of the Ba adatoms at 300 K. In Fig. 3 we have also shown the distribution $n(x)$ obtained after heating of the crystal at 400 K ($t = 2.5 \text{ min}$). To record this distribution the crystal was cooled to 77 K. Let us consider the fact that the $n(x)$ curve is highly asymmetric with respect to the point of intersection with the edge of the initial distribution and that structure is distinctly observed in it—regions with reduced concentration gradient in the vicinity of $n \approx 1.2 \times 10^{14}$ and $3.5 \times 10^{14} \text{ cm}^{-2}$ (degree of coverage respectively 0.2 and 0.6). This shape of the distribution indicates a strong dependence of the diffusion coefficient D on n .

Figure 4 shows the dependence $D(n)$ obtained at 400 K by the method described above. It is evident that D , as

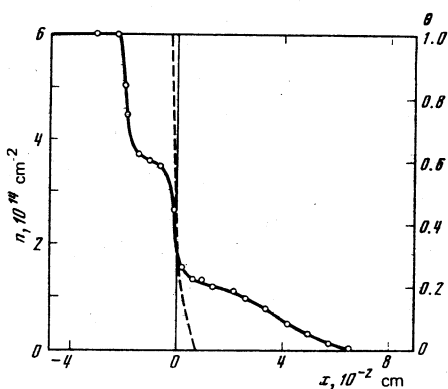


FIG. 3. Distribution of Ba atoms after heating at $T = 400 \text{ K}$ for 2.5 min. The dashed line is the initial distribution. θ is the degree of coverage.

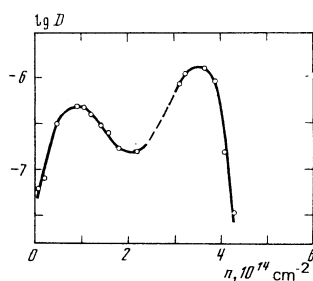


FIG. 4. Dependence of D ($\text{cm}^2 \text{ sec}^{-1}$) on n at $T = 400 \text{ K}$.

expected, has maximum values at concentrations corresponding to the plateau segments in the $n(x)$ distributions. Within the range 0 to 1 monolayer, D varies by more than two orders of magnitude.

Figure 5 shows Arrhenius plots constructed from the data on the basis of Eq. (2). The experimental points lie on straight lines with a small spread. The values of $E_d(n)$ found from the slope of these straight lines shown in Fig. 6. Note that in the region $n \approx (2.2-3.2) \times 10^{14} \text{ cm}^{-2}$ we were not able to determine D and E_d , as a result of the fact that in this concentration interval the work function depends very weakly on n and the method used has poor accuracy.

In the case when the coverage in the initial film with a sharp boundary exceeds a monolayer, we also observe in the $n(x)$ curves portions with reduced concentration gradient at $n \approx 1.2 \times 10^{14}$ and $3.5 \times 10^{14} \text{ cm}^{-2}$ (Fig. 7). Note that the step at $n = 3.5 \times 10^{14} \text{ cm}^{-2}$ is found in all cases to be more clearly expressed. In the presence on the substrate of a pedestal ($1.2 \times 10^{14} < n_p < 3.5 \times 10^{14} \text{ cm}^{-2}$) only one step at $n = 3.5 \times 10^{14} \text{ cm}^{-2}$ is formed in the $n(x)$ distributions. An example of such a distribution was given in our earlier study.⁹ We emphasize that in the initially uncovered portion of the surface it is not possible to obtain a monolayer covering by diffusion of barium from a thick film with a sharp boundary (at least, at distances from the initial boundary which can be recorded within the resolution of our method). Attempts to achieve this by increasing the temperature leads only to a rapid evaporation of barium from the multilayer covering.

We also carried out several experiments aimed at study of the anisotropy of barium diffusion. Here the initial covering was evaporated in the form of a circle of diameter $100 \mu\text{m}$. As a result of diffusion the lines of constant concentration in the $n(x, y)$ distribution acquire the shape of ellipses with a major semi-axis oriented along a direction of the $[100]$ type. The ratio of the axes of the ellipses was 2-4.

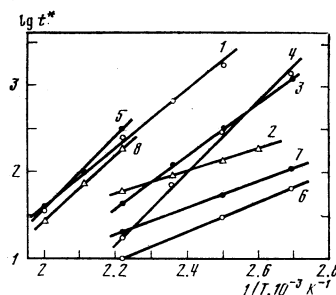


FIG. 5. Arrhenius lines for various n (10^{14} cm^{-2}): 1—0.1, 2—1, 3—1.45, 4—1.7, 5—1.9, 6—3.3, 7—3.9, 8—4.3; t^* is in seconds.

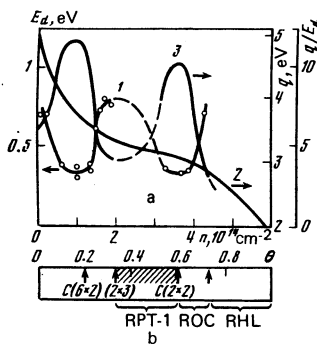


FIG. 6. a) Diffusion activation energy E_d (1), heat of adsorption q (2), and the ratio q/E_d (3) as functions of n . b) Structure diagram for Ba films on Mo(011) (Ref. 3): RPT-1—region of phase transition of the first type (two-dimensional condensation), ROC—region of one-dimensional compression of the $c(2 \times 2)$ structure in the $[100]$ direction, RHL—region of hexagonal lattices with smoothly decreasing period as θ increases (for models of the structures see Fig. 8 below).

DISCUSSION OF RESULTS

The diffusion distributions obtained differ radically from those predicted by diffusion theory for noninteracting particles. We recall in this connection the basic information on interaction of barium atoms on the (011) face of molybdenum, obtained in study of the atomic structure of barium films and the concentration dependence of the heat of adsorption.³

With increase of the concentration, the barium atoms successively form more and more dense two-dimensional lattices (Fig. 8). The increase in density of the film in the interval $\theta = 0-1$ is accompanied by a strong decrease—almost by a factor of three—in the heat of adsorption (Fig. 6, curve 2). These facts are naturally explained by the presence of a repulsive interaction between the adatoms, due to the strong polar nature of the adsorption bond. However, the dipole moment of the bond and the interaction energy of the barium adatoms vary in a complicated way with increase of the degree of coverage. In particular, this is indicated by the existence at intermediate θ values of a broad region of two-dimensional condensation (Fig. 6b)—a phase transition of the first kind from $\theta = 0.33$ [a (2×3) lattice with vacancies] to $\theta = 0.6$ [a $c(2 \times 2)$ lattice]. Breakup of the film into phases with different density becomes energetically advantageous at some critical coverage as a consequence of the mutual depolarization of the adatoms with increase of θ .¹¹ This can be interpreted as the result of appearance in the film of effective attractive for-

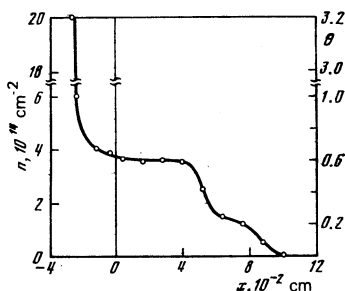


FIG. 7. The dependence $n(x)$ after heating of a thick Ba film at $T = 400$ K. The initial coverage is $\theta = 3.2$.

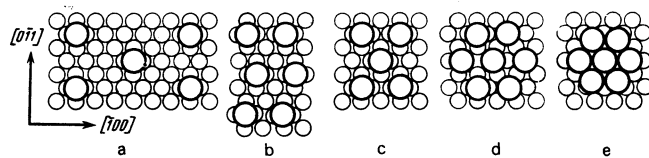


FIG. 8. Two-dimensional lattices of Ba atoms on Mo (Ref. 3): a— $c(6 \times 2)$, b— (2×3) , c— $c(2 \times 2)$, d—hexagonal structure registered with substrate along $[0\bar{1}1]$ axis; e—hexagonal structure of dense monolayer.

ces which result in condensation of atoms representing an excess over the critical coverage into a denser phase.

When we compare these data and the results obtained in the present work we can state that all of the basic steps of structural transformations in the film appear in the characteristics of surface diffusion. The steps in the distributions $n(x)$ and the extrema in the functions $D(n)$ and $E_d(n)$ are found at concentrations corresponding to $c(6 \times 2)$ and $c(2 \times 2)$ lattices. This correlation exists in spite of the fact that at the temperatures at which diffusion has been studied, long-range order has not been observed in these lattices, although short-range order is preserved.³

Let us consider first the diffusion from a thick layer (Fig. 7). As has been mentioned above, in this case the phase $\theta = 0.6$ is actually propagated over the surface [experimentally it has been possible to obtain well shaped steps of this phase extending for distances ~ 1 mm (Ref. 9)]. This nature of the distributions can be explained by the following special properties of the $\theta = 0.6$ phase. First, it is a two-dimensional condensate which can be in local equilibrium with a less dense coverage immediately adjacent to it in the diffusion distribution. Second, the diffusion coefficient at $\theta = 0.6$ has its greatest value (Fig. 4). As a result of this all the barium moving from the left from the initial covering is rapidly transported in the $\theta = 0.6$ step, so that the concentration gradient within that step remains insignificant.

Regarding the causes of the high mobility of barium atoms at $\theta = 0.6$ we can present the following reasoning. The phase $\theta = 0.6$ is unstressed, since it is in equilibrium with the less dense phase $\theta = 0.33$. However, excess atoms arriving in the condensate produce in it a strong lateral compression. This is indicated, in particular, by the rapid decrease of the heat of adsorption on increase of the coverage from $\theta = 0.6$ to $\theta = 1$ (Fig. 6). We can suggest that on introduction of an excess atom an interstitial configuration of some type is formed. It is well known^{12,13} that such configurations—for example, crowdions in a system with long-range order—have a high mobility. Displacement of such a configuration to the boundary of the $\theta = 0.6$ phase permits removal of the stress which has arisen in the film and simultaneously leads to a further advance of the boundary of this phase along the surface (Fig. 9a).

Thus, the unrolling-carpet mechanism, in which a close-packed monolayer propagates along the surface¹⁴ (Fig. 9b), is not realized for the system investigated. This mechanism is characteristic of systems with an

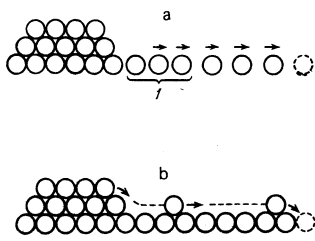


FIG. 9. a) Proposed mechanism of diffusion from a thick layer according to the distributions observed in the present work and according to the data of Refs. 2, 3, and 9; 1—interstitial configuration of adatoms, b) unrolling-carpet mechanism according to Ref. 14.

attractive interaction of adsorbed atoms in the first monolayer.⁵ With a repulsive action of the adatoms the dense first monolayer is stressed (under compression) and formation of this layer at the leading edge of the diffusion distribution is energetically disadvantageous.

Let us consider now in more detail the variation of the diffusion parameters over the entire region of submonolayer coverage $\theta < 1$.

The region $\theta < 0.2$ corresponds to the first falling portion of the $E_d(\theta)$ curve. Similar behavior of E_d at small θ has been observed by Schmidt and Gomer¹⁵ in diffusion of potassium over the surface of a tungsten tip made up of a set of different faces, and also in diffusion of thorium on the (113) face of tungsten.² Schmidt and Gomer¹⁵ explain the decrease of E_d in regions of low θ by successive filling of adsorption sites with lower and lower binding energy. The inhomogeneity of the surface in adsorption may be due either to the presence of defects or to the crystal structure of a perfect face. In our case the concentration of adatoms in the θ interval considered is an order of magnitude less than the concentration of holes on the (011) face of molybdenum (Fig. 8), and therefore the intrinsic nonuniformity of the surface in this case should not yet affect the dependence $E_d(\theta)$. Above we have presented facts indicating the existence in the investigated substrate of a certain number of defects. However, it is hard to assume that it could amount to $\sim 10^{14}$ cm⁻² (but the decrease of E_d in the first falling portion occurs up to $n = 1.2 \times 10^{14}$ cm⁻²). It is evident that under this assumption there could not be formed a regular lattice of adatoms $c(6 \times 2)$ whose periods are multiples of the periods of the perfect (011) face of molybdenum. Thus, we reach the conclusion that the dependence $E_d(\theta)$ over practically the entire coverage region studied—except perhaps for a very narrow region as $\theta \rightarrow 0$ —is due to the structure of the Mo(011) face itself and to the interaction of the diffusing barium atoms with each other.

It is understandable that for a rigorous interpretation of the dependence $E_d(\theta)$ it would be necessary, knowing the interaction law, to carry out detailed calculations of E_d for various possible diffusion mechanisms. Here we confine ourselves to a preliminary qualitative analysis of the data.

The rapid decrease of the heat of adsorption at small θ , which indicates a strong repulsion of the adatoms,

is accompanied by a rapid decrease of E_d . The first minimum of E_d corresponds to the coverage at which the $c(6 \times 2)$ lattice is formed (long-range order exists in it³ below $T_c = 100$ K). In the region $0.2 < \theta < 0.33$ the heat of adsorption continues to decrease, although much more slowly, while E_d rises significantly (the increase of E_d with θ has been observed also for thorium and potassium on tungsten^{2,15,16}). This may indicate a change in the diffusion mechanism. This will hardly be a vacancy mechanism, since there are still a large number of vacant sites on the surface. We note the fact that the θ region under discussion is precritical—it occurs directly before the region of two-dimensional condensation. Modeling by the method of molecular dynamics^{17,18} shows that under these conditions one can have a cluster mechanism of diffusion, in which a group of a small number of atoms migrates as a whole in a less dense background.

As we have already remarked, we were unable to obtain data on E_d in the region of two-dimensional condensation. The value of E_d should apparently be rather large in this region, since the limiting stage of the redistribution of adsorbate over the surface will be the stripping of adatoms from the islands of two-dimensional condensate. This requires in addition the expenditure of the energy of two-dimensional sublimation.¹⁹

Near $\theta = 0.6$, where the two-dimensional condensate completely covers the entire surface, E_d passes through a second minimum. Possible causes of the high mobility of the adatoms at this coverage have been discussed above.

Further increase in density of the film ($0.6 < \theta < 1$) leads to a destruction of its structural registry with the substrate,³ to a new rapid decrease of heat of adsorption, and to an increase of E_d . As a consequence of the decrease of the ratio q/E_d (Fig. 6a, curve 3) the mean free path for diffusion of atoms before their evaporation gradually is shortened. For this reason it is practically impossible to cover a macroscopic portion of the surface with a multilayer film of barium by diffusion from a thick layer: at high temperature the adatoms rapidly evaporate, and at low temperatures the rate of diffusion is too small.

Thus, the dependence $E_d(\theta)$ obtained reflects the complex nature of the change of the interaction in the barium film with increase of the degree of coverage. This is illustrated also by the following observation. As has been shown by Bowker and King,⁷ with repulsion of the adatoms the point of intersection of the diffusion distribution with the line marking the initial boundary of the film with $\theta_{\max} = 1$ should lie above $\theta = 0.5$, while for attraction it should lie below $\theta = 0.5$. Although there is no doubt regarding the existence of repulsive interaction between barium adatoms, for this film the indicated point lies below $\theta = 0.5$ (Fig. 3). This is apparently due to the existence of the region of two-dimensional condensation, in which effective forces of attraction are acting.

In conclusion let us mention the question of the anisotropy of diffusion of barium on Mo(011). Butz and Wagner⁵ in study of the diffusion of palladium on W(011)

established that palladium diffuses with increased velocity along steps in the surface. It is still unclear to what degree the distributions $n(x, y)$ obtained by us may be due to the influence of steps and in general what is the nature of the influence of the steps on the diffusion of adatoms with attractive and repulsive forces. This question requires special study.

CONCLUSION

The results presented show that in the characteristics of surface diffusion there appear distinctly all the features of interaction of the particles in the adsorbed film, which appear also in the formation of definite two-dimensional lattices of adatoms. The step-like distributions of concentration of diffusing particles on the surface are similar to the diffusion distribution in a volume, obtained on formation of definite phases in the diffusion process.²⁰

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