Processes of Nucleation of Amorphous As-S Films at Condensation on Carbon Substrates

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Initial stages of vacuum condensation of As-S amorphous films on carbon substrates at the stages of nucleation and formation of condensed phase islands are studied using the electron microscopy technique. Two stages of the processes of critical nuclei formation were found: intense primary nucleation and slow secondary nucleation that occurs in parallel with the growth of the formed nuclei. The sizes of the critical nuclei of the amorphous As-S films, the primary nuclei density, the size distribution of the condensed phase islands at the moment of the nucleation processes termination and the mean diffusion length for different atomic particles of the As-S vapor phase on the substrate surface were determined. The results obtained show that at room condensation temperatures most of various multi atomic particles of the As_nS_m vapor have rather high diffusion mobility over the carbon substrate surface. Therefore, when simulating the condensation processes, the As-S materials could be considered as a non-equilibrium two-dimensional gas with an extremely complicated chemical composition.

Keywords: Condensation mechanism, Islands films, Amorphous films, Chalogenides.

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1. INTRODUCTION

Processes of formation of condensed films of different-material have been extensively studied and analyzed in the second half of the last century. The relevant results were published in thousands of original experimental and theoretical papers, most of them being devoted to studies of condensation of crystalline layers and reviewed in well-known monographs [1-3]. The character of processes of condensation on the substrate is described by such basic mechanisms as the Vollmer-Weber nucleation mode, the Frank-van der Merve growth mode with a strong interaction of the condensate with the substrate and the two-dimensional Stranski-Krastanov growth mode [4].

However, the suggested models often fail to explain even qualitatively many facts revealed in the experiments on the vacuum film condensation. This is due to the fact that the above mechanisms refer to the low oversaturation of vapor flows and the ideal condition of the substrate surface. At such idealized conditions, the surface density of atoms condensed on the substrate is determined solely by the incoming vapor flow density and the energy of desorption of atoms from the substrate surface. Both above parameters are assumed to be independent of time and constant for all condensed vapor particles. Such stationary conditions make a good physical approximation only for the condensation processes described by a chain of separate sequential quasi-equilibrium acts of vapor particle arrival at the substrate surface. In reality, the condensation processes occur in a much more complicated manner with considerable deviations from the thermodynamical equilibrium conditions. Another shortcoming of the above theories is the use of a number of such macroparameters as the surface tension, density and surface energy that are not applicable to describe a system consisting of several or several dozens of atoms.

Within recent years, the structure of As-S thin films was intensely studied to understand the mechanisms of structural transformation in these materials under laser irradiation [5, 6]. Still, the physical processes responsible for various photo-stimulated changes in As-S films remain unclear. In our opinion, this problem can be solved only by taking into account all physicochemical processes of the formation of the As-S amorphous film network at the vacuum condensation. The three above condensation mechanisms are even less adequate for real technological processes of amorphous chalcogenide film deposition, including the As-S materials, where the vapor flow condensation conditions and processes often are more complicated. The main of them are: (1) strong oversaturations, for which the criticalsize nuclei can contain only several atoms, (2) prior to coalescence, the size of the most of the nuclei of the condensate on the substrate has not reached the critical nucleus size yet, (3) transient character of the amorphous film condensation and growth processes, especially at their initial stages, (4) effect of the existing nuclei and their growth processes on the concentration of atoms adsorbed on the surface, (5) specific features of condensation processes for multiatomic clusters of the vapor flow as compared to the behavior of

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individual adsorbed atoms. As of today, microkinetic theories [7] more adequately reproduce the real processes of the film condensation in vacuum. They are based on the use of a system of kinetic equations that describe the behavior of all atomic particles condensed on the surface at the initial film nucleation and growth stages. In this case, parameters are introduced into the equation that characterize the state of the condensation surface, in particular, concentration of the specific active centers at which accommodation of the particles coming from the vapor occurs more intensely. Still, the microkinetic theories encounter a number of difficulties while describing the processes of thin film condensation, especially for amorphous films. The main of them are as follows: (1) the need to introduce instantaneous accommodation of the vapor flow atoms on the active centers of the substrate surface; (2) kinetic equations have simple solutions only under the condition that the number of atoms localized on the active centers exceeds that of the atoms accommodated on other areas of the substrate; (3) the mean diffusion distance for atoms on the surface is assumed to considerably exceed the distances between the active centers; (4) the presence of particles of different-type in the vapor flow strongly increases the number of kinetic equations to be solved to study the compound thin film condensation.

Analysis of the state of art on the investigation of amorphous film condensation processes shows that, as of today, a lot of problems still remain unsolved. Here both the macrothermodynamical and microkinetic theoretical methods of studying the nucleation and cluster growth on the substrate surface meet a number of principal difficulties. This especially concerns the processes of deposition of compound As-S materials, for which the vapor flow can contain various atomic complexes and clusters [8]. Most of approximations within the theoretical models of such processes do not comply fully with real conditions or are reproduced only within narrow intervals of variation of technological parameters of condensation. A constrained limitation of the number of differential equations in microkinetic approaches excludes the possibility of a sufficiently exact calculation of the main parameters of the vacuum deposition of thin layers. All the above factors stipulate a very small number of theoretical and experimental studies of the amorphous film condensation processes, especially for compound substances.

The above difficulties are also revealed in computer simulation of vacuum condensation of amorphous As-S films. The possibilities of average modern computers restrict the size of the model systems by a few thousand of atoms. When condensation of amorphous layers is studied, such restriction allows one to investigate only the processes of formation of separate condensed phase nuclei with their further evolution to nanometer-sized islands, specific for the coalescence stage of the deposited film. However, even for such utterly simplified simulation conditions, one should take into account several important parameters of the condensation process, namely the critical nuclei size, their surface density and the energy characteristics of adsorption and desorption of different vapor phase particles on typical substrates. In this work, these parameters are determined for the materials of As-S system based on the experimental electron microscopic studies of the processes of critical nuclei formation and island film growth on carbon substrates.

2. EXPERIMENTAL

The samples under study were produced using traditional methods of simple thermal evaporation of initial glasses with the vapor flow deposition onto substrates in vacuum. This method is most widely used for the studies of As-S thin films. Technological operations were carried out at a conventional VUP-5 vacuum deposition setup equipped with different facilities for changing substrates and controlling the deposition process parameters. The intrachamber facilities allowed sequential independent film deposition onto six separate substrates to be performed using two different evaporators within one technological cycle without vacuum deterioration. To ensure identical condensation conditions for all the six samples, the substrates were placed at a carousel providing the same conditions of the vapor flow.

Supporting layers of ultrathin (~10 nm) carbon films were used to enable sample preparation in a form of the separate nuclei and to carry out their structural studies. All of them have highly homogeneous intrinsic microstructure and provide an extremely low contribution to the electron microscopic image contrast [9].

The technological cycle of production of samples under study included a series of certain operations. (1) Preparation of the equipment under the bell-jar and the vacuum apparatus. (2) Deposition of a supporting thin carbon layer onto six NaCl substrates by conventional evaporation technique using carbon rods; this film thickness was controlled by the spraying time at the given technological conditions. (3) Starting the chalcogenide material deposition onto all substrates. (4) After each 2-10 seconds one of the six substrates was sequentially shielded by special shutter screens. This means that the duration of condensation was 2-12 seconds for the first substrate and 10-60 seconds for the last (sixth) one. Due to this, we succeeded to obtain (within the same technological cycle) films at different stages of nucleation, island formation and merging. (5) After terminating the condensation process, the films were kept in a high vacuum for 15-20 minutes and then removed from under the bell-jar.

Evaporators with direct electric-current heating were used for evaporation of the chalcogenides. Tantalum or niobium foil was used as the evaporator material. A Pt/Pt-Rh thermocouple was mounted in a special corrugation on the evaporator housing and was used to measure the initial chalcogenide evaporation temperature. A rectangular parallelepiped-shaped effusion cell was the main evaporator types. As our studies have shown, such closed effusion evaporators provide technological conditions of rather high quality island-type amorphous As-S films. to produce Averaged integral geometric thicknesses of the islandtype films produced at different stages of their formation were 1-50 nm. The average geometric thickness was controlled by setting the mass of the substance to be evaporated. The thickness obtained were calibrated by an ultrasonic method. In this case,

an accurately weighed amount of substance was evaporated at fixed deposition geometry to produce a 20-30 nm thick test film. After measuring this test film thickness by the ultrasonic method, we determined the mass of the material to be evaporated, required to provide the deposition of samples with smaller thicknesses in the same geometry. In this case the specific density of the condensate was taken equal to that of the initial glass.

The island-like films produced were studied by transmission electronic microscopy using an EMMA-4 apparatus and standard methods. All the images were detected in sharp focusing mode. Some experiments were carried out using a JEM-2010 electron microscope (from JEOL) equipped with a filter for inelastically scattered electrons. It was found in the course of electron-microscopic experiments that few-nanometer-sized islands of the chalcogenide material provide a quite good contrast against the supporting carbon film background. This is due to the fact that the total electron scattering cross section for much heavier As and S atoms is almost by an order of magnitude larger than that for the C atoms [10].

3. RESULTS AND DISCUSSION

We studied As_nS_m cluster adsorption on the carbon substrate surface by means of the computer calculation using the ab initio quantum-mechanical methods [11]. These results show that it is at the stage of formation of a two-dimensional adsorbed layer when the effect of the As-S vapor phase composition on the kinetics of its condensation is revealed most vividly. This is due to the fact that different multi-atom complexes being thermodynamically stable in the vapor phase may behave differently on the substrate surface, thus making the general pattern of the condensate formation more complicated. However, as the quantum-mechanical calculations show, the atoms of the most of the As-S vapor phase clusters have no free valences [11]. Therefore, at the initial stages of deposition onto the substrate, different As_nS_m clusters contact with the surface by means of at least five atoms bound to it solely by weak molecular forces [12], their value being close to that of the forces of interaction of separate atoms with the condensation surface. Meanwhile, the results of Ref. [7] show that the influence of different multi-atom clusters on the nucleation processes in the course of condensation is similar if the activation energy of their surface diffusion is close to that for separate atoms. Thus, the results of the ab initio calculations and computer simulation show that at the initial condensation stages almost all the particles of the As-S vapor flow have a large mobility over the surface.

At the condensation of amorphous As-S films, the substrate temperature is kept quite low. At such conditions, the relaxation times of the excessive energy of the adsorbed particles are much larger than the period of thermal atomic vibrations. Therefore, at the initial condensation stage, all the As_nS_m particles have a certain diffusion mobility across the surface and migrate intensely from one potential well of the substrate energy landscape to another [12]. The main driving force of such surface migration of the adsorbed vapor particles

is their intrinsic thermal kinetic energy. The specific features of this diffusion mobility determine to a great extent the character of the film formation and its structure. After the thermal energy transfer from the $\mathrm{As_nS_m}$ clusters to the substrate, they are bound to it by strong covalent bonds and almost lose their mobility.

Thus, when modeling the initial condensation stages, the system of the mobile As_nS_m particles on the substrate surface can be treated as a specific and a very complex (from the point of view of chemical composition) non-equilibrium "two-dimensional gas". Different particles of this gas reaching the substrate from the vapor have a certain condensation temperature T_c determined by their average kinetic energy. During their lifetime, these particles lose gradually their energy and migration ability, reach the condition of thermal equilibrium with the substrate at the temperature T_S and are chemically absorbed on it. As a result, they are rapidly fixed on the substrate or on a growing condensate island producing a spectrum of different-type chemical forces of both physical and chemical nature. New atomic complexes of the two-dimensional gas from the vapor flow substitute the condensed particles. Due to such processes at low T_S , nuclei of the condensed amorphous film can be formed from the non-equilibrium two dimensional gas at the substrate surface. Note that in case of the As-S materials separate multiatomic vapor particles may also become individual stable nuclei of condensate.

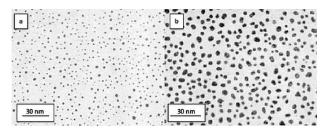


Fig. 1 – Electron microscopic images of the initial stage of nucleation of the amorphous $As_{20}S_{80}$ (a) and $As_{40}S_{60}$ (b) films. The condensation conditions: $T_S = 300$ K, $v_{\rm K} = 0.2$ nm/s, condensation time -4 s (a), 9 s (b)

Calculations [7] show that dissociation of the existing nuclei clearly affects nucleation processes only for those systems, in which the activation energy of the adsorbed particles surface diffusion is close to the dissociation energy of the initial nuclei. According to the quantum-mechanical calculation data for As-S [11], this condition in most cases does not hold true. Therefore, we assume that dissociation of the already formed nuclei is not significant. Our electronmicroscopic studies have shown that at low deposition rates most of the amorphous films of the As-S system indeed condense on different substrates according to the nucleation mechanism (Fig. 1). In accordance with the statistical approach [13], a certain time passes from the condensation onset, during which the required density of the two-dimensional gas on the substrate surface is achieved. No condensate nuclei are formed during this period of time. For the room temperature of the substrate and low condensation rates, this time interval can reach several seconds. According to our experiments, at $T_S = 300$ K and $v_R = 0.2 \cdot 0.5$ nm/s, we

using the electron microscopy technique, failed to find amorphous As-S film nuclei on the substrate when the condensation time was less than 2-5 s. Thus, during this period of time a sufficiently dense two-dimensional gas of the As-S system vapor flow particles is formed on the surface with the ability to produce a system of critical nuclei of the amorphous phase and grow to the size which can be reliably detected in electron microscopic images. This situation is achieved when the condensate thickness reaches the integral value of 0.5-1 nm. Probably, at this stage the integral film thickness includes both the formed critical nuclei and the two-dimensional condensed gas. According to our electron microscopy data, the density of the critical nuclei on the carbon substrate exceeds 0.05/nm² whereas the mean diameter of some nuclei does not exceed 2 nm (Fig. 1).

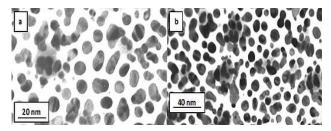


Fig. 2 – Electron microscopic images of the condensed phase islands growth and secondary nucleation for the amorphous $As_{10}S_{90}$ (a) and $As_{30}S_{70}$ (b) films. The condensation conditions are as follows: $T_S = 400$ K, $v_{\kappa} = 0.5$ nm/s, condensation time – 7 s (a), 10 s (b)

Here we analyze the experimental results for the simplest case of homogeneous substrate surface. Adsorption of a certain vapor particle on any point of this surface has equal probability. In this case, the adsorption energy E_{ad} of a given particle at all points of the surface will be the same as well. Thus, we do not take into account the real structure of the substrate surface. This approximation will work well at low concentrations of defect centers on the substrate surface. In accordance with the conclusions of the statistical theory, within the framework of the assumed condensation surface model, at quite high substrate temperatures and low vapor oversaturation degrees, stable condensed phase nuclei should contain more than 20 atoms. The critical As-S nuclei size, which we observe experimentally, agrees fairly well with the relevant theoretical estimations.

With decreasing substrate temperature and increasing condensation rate/oversaturation degree, the critical nuclei size decreases gradually within the framework of statistical theory. In the limiting case, such a nucleus can be formed even by two separate atoms forming a chemical bond due to an accidental meeting. However, amorphous nuclei smaller than 2 nm can hardly be found on the carbon substrate using conventional transmission electron microscopy methods. Therefore, we could not find a trend of decreasing the size of the critical amorphous As-S film nuclei with the above variation of the technological conditions.

Thus, both the theoretical analysis and the experimental studies show that, at typical conditions of vacuum condensation, the initial stage of the condensed As-

S film formation includes formation of a system of critical-size nuclei on the substrate surface from the oversaturated vapor. In this case the size of the critical nuclei is less than 2 nm and is determined by the balance of their energies as compared to the energy of separate vapor particles that form it.

Subsequently, the critical nuclei formed start to grow intensely, being transformed with time into stable islands of a newly formed condensed phase (Fig. 2). Nuclei with the size below the critical value are energetically unstable. They can decay, if during their lifetime they are not transformed into critical-size ones, or, otherwise, they form new critical nuclei. Meanwhile, as can be seen from Figs. 1 and 2, the nuclei and the islands are distributed rather uniformly on the condensation surface and clearly separated from each other. Thus, during their stay in the two-dimensional gas, the vapor flow particles are able to move across the surface to substantial distances l_D prior to be thermally accommodated. We may take the l_D value approximately equal to half of the mean distance between the neighboring critical nuclei or the islands formed. Simple calculations for structures similar to those shown in Fig. 1 give $l_D \sim 4-5$ nm. This agrees well with the theoretically calculated diffusion path length for different particles of the As-S vapor [12].

An essential question arises: why the As-S vapor particles having energy sufficient to migrate across the surface for approximately a dozen of nanometers fail to ensure their binding into an ordered crystal lattice while for this they have to pass only few angstroms? In our opinion, the reason is a complex intrinsic structure of most of the particles of the vapor flow reaching the substrate and strongly non-equilibrium processes of these particles binding into the atomic network of the film

After the formation of critical-size nuclei (primary nucleation), the flow of the absorbed particles from the surrounding substrate area sharply increases. This surrounding area can be presented as a drain circle around each nucleus. The diameter of this circle for different nuclei can depend on the mass composition of the vapor flow and its condensation rate [13]. In our experiments, such dependences were revealed as a decrease of the mean circle (diameter) by 20-30 % with ν_{κ} increasing from ~0,2 nm/s to ~2 nm/s and at the condensation of the amorphous films with larger As content (Figs. 1 and 2). As a result of the rapid drain of vapor particles from the circle area with further condensation the concentration of the adsorbed vapor particles here remains lower as compared to other areas of the substrate. Beyond the drain ring, the total flow of particles toward the nucleus is much smaller and the particle concentration is determined mainly by the degree of the vapor oversaturation on the substrate. In these areas, the adsorbed particles are accumulated, and new critical nuclei of the condensed phase are actively formed (secondary nucleation). We often noticed such processes in the electron microscopic images (Figs. 1 and 2). Note also that, according to Ref. [13], the higher is the condensation rate, the sharper is the concentration drop of the adsorbed particles in the surface regions in the center and beyond the outer boundary of the drain circle.

Thus, each stably growing amorphous film nucleus has its own drain circle. If the adsorbed vapor particles reach this area, they drift toward the relevant nucleus providing its growth. With increasing nucleus size, the drain area increases. When the nuclei grow up to a certain limiting size, the secondary nucleation at the substrate becomes impossible and stops. Such processes determine the maximal number of stable nuclei and condensed phase islands on the substrate surface that in case of the amorphous As-S films reached ~ (0.07-0.08)/nm⁻² on the average. Comparing this value with the critical nuclei concentration at the initial stages of the amorphous As-S film condensation ($\sim 0.05 \text{ nm}^{-2}$), one can state that the number of islands produced due to the secondary nucleation is less than due to the primary nucleation processes. At the terminating stage of the nucleation on the carbon substrate surface, the amorphous As-S film islands were 2-15 nm in size (Fig. 2).

The electron microscopic studies do not allow one to find a direct role of the multiatomic vapor phase clusters in the nucleation of the amorphous As-S film. However, in our opinion, the less mobile vapor phase clusters consisting of 6-10 atoms will be the centers of formation of many critical nuclei [1]. The role of such clusters in the nucleation of the As-S film will be determined by their structure as well as physical and chemical parameters of their deposition onto the substrate. Time and energy required for certain atoms to be bound are the basic parameters that determine the regularities of the thin film structure formation, including their trend to the formation of an amorphous state. The above analysis shows that in case of the amorphous As-S films this condition should be as well extended to multiatomic vapor complexes.

Large-scale As-S vapor complexes posses an intrinsic structure [11]. Their specific features shall definitely affect the character of behavior of these complexes on the substrate in case of a contact with the latter. At the stage of condensation, they will keep turning around and rolling over until the moment when their most energetically active "faces" contact the substrate surface. At the final stage of binding, the turnovers and slight "crawling" of the particle that ensure its maximal energy of interaction with the substrate surface can also be important. With decreasing size of the multiatomic complexes, their surface mobility at the substrate will considerably increase, reaching its maximal value for separate atoms. At deep overcooling, is the stochastic random process of different vapor particles binding with the emerging condensation centers and the growing surface that leads to the formation of the structures in the As-S amorphous films that differ essentially from the massive glass structure and from other types of amorphous state for these substances.

At the next condensation stage the critical nuclei, gradually increasing in size, are transformed into condensed phase islands. The processes of formation of the final amorphous As-S film and the specific features of its structure depend on the above nuclei concentration, size, shape and character of interaction with one another. Another important parameters are the chemical composition of the substrate surface layer (taking into account the presence of the impurity elements in

it), the type of chemical bonds between the atoms of the substrate surface layer as well as nano- and micro-scale surface topography. Probably, in the case of amorphous As-S films, their effect is somehow masked, but, nevertheless, these factors are actively revealed at the initial film growth stages and affect the final structure of the condensates and their properties [14].

The above analysis of the amorphous As-S film nucleation taking into account solely the parameters of certain vapor particles on the surface is quite simplified since in these processes the main role is played by the binding energies of the adsorbed particles with the surface. Formation of a more thermodynamically instable amorphous phase in the nuclei is mostly due to kinetic reasons, not the activation energies E_{ac} of formation of different-phase nuclei on the substrate. In other words, formation of a more thermodynamically instable, but kinetically beneficial amorphous phase of the As-S materials results from the fact that the activation energy of such process at chemically conservative self-assembling determines not the nucleation processes, but only their rate. Therefore, it is not surprising that many properties of amorphous and polycrystalline As-S films are practically independent of the substrate type. For a more detailed understanding of the nucleation and film growth processes in such materials, one has to draw considerable attention to the behavior of separate atoms as well as multiatomic vapor phase complexes and clusters taking into account the character and energy of the bonds that can be formed in the process of condensation [15].

Afterwards, the condensate nuclei grow both due to the new vapor particle inflow from the two-dimensional gas and at the expense of the particles that "fall" directly from the vapor flow onto the nuclei. At the initial stages of nucleation and nuclei transformation into the small islands, one may neglect the second channel of the condensate material inflow. However, with the increasing island size, the role of this channel gradually increases and it becomes the main one after the separate islands merging into the solid film.

The above results were obtained in the approximation of an ideal homogeneous condensation surface. At realistic technological conditions, the substrate surface is in most cases microheterogeneous, i.e. contains defect areas of different nature. In these areas, the parameters of the vapor particle absorption and their two-dimensional migration can differ essentially. As a rule, the nucleation processes at the defect centers occur more intensely. Therefore, at a high concentration of defect centers, the density and size of the amorphous As-S film nuclei on the substrate surface will be determined mainly by the condensation processes with participation of various defect areas. In general, nucleation can occur both on homogeneous and defect surface areas.

4. CONCLUSIONS

According to the mass-spectrometric data, about 20 different As_nS_m (n, m < 6) atomic complexes exist in the As-S vapor. As the experiments show, at the initial stages of condensation of these materials with small rates of 0.1-1 nm/s, separate amorphous critical nuclei

located at considerable distances from each other are formed on the carbon substrate surface from the vapor phase. This indicates that at room condensation temperatures the majority of multiatomic particles of the $As_{n}S_{m}$ vapor have large diffusion mobility on the surface and intensely migrate across it. Thus, when simulating the condensation processes, the As-S materials on the substrate surface can be treated as a heterogeneous "two-dimensional" gas with extremely complicated chemical composition. In this case, the stage of nuclei formation and growth includes two phases.

At the first phase, the mass formation of the critical nuclei occurs. This stage lasts for 3-7 s after the condensation onset and corresponds to the case of reaching the integral thickness of the condensate of 0.5-1.0 nm. The density of the critical nuclei formed at this phase on the carbon substrate exceeds $0.05/\mathrm{nm}^2$, whereas their average diameter does not exceed 2 nm. The initial nuclei are distributed quite uniformly over the substrate surface with the average distance between them of about 10 nm.

At the second phase, the critical amorphous nuclei produced grow slowly with transformation into condensed-phase islands. Meanwhile, an active formation of new critical nuclei, i.e. the secondary nucleation, starts on the substrate areas free of the primary nuclei. In this case, the secondary critical nuclei are formed only at large distances from the existing amorphous film islands. Such processes show that around each stable growing nucleus on the condensation surface there is a circle of intense drain of adsorbed vapor particles to this nucleus. The density of heterogeneous non-equilibrium two-dimensional gas within these circles is substantially less than in other substrate surface areas. Outside the above drain circles, the density of the two-dimensional gas is determined mainly by the vapor oversaturation degree that favors the formation of new secondary critical nuclei in these areas. The distances between the primary and the secondary nuclei correspond to the mean diffusion path length for different As_nS_m particles on the condensation surface (below 4-5 nm).

After the concentration of the growing island reaches a certain limit, the secondary nucleation on the substrate becomes impossible and ceases. This stage corresponds to the maximal number of stable nuclei and islands of the condensed As-S material phase on the substrate surface, equal on the average to $\sim 0.1/\text{nm}^{-2}$. Thus, the amorphous As-S islands are mostly produced due to the primary nucleation processes. At the stage of the nucleation process termination on the carbon substrate surface, the sizes of the condensed phase islands lie within a broad range from 2 to 15 nm. The integral film thickness is about 10 nm.

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