Initial stages of diffusion and phase formation in Sc/Si layered systems

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Kinetics of phase formation Sc/Si multilayers and Si/Sc/Si three-layers within the temperature range of 130–400°C has been studied by cross-sectional transmission electron microscopy and small-angle X-ray reflectometry. Growth of ScSi silicide governed by diffusion kinetics was observed at all temperatures. Two growth stages of nonparabolic and parabolic behavior were revealed. Initial nonparabolic stage is characterized by higher diffusion coefficient, which decreases during annealing in 20 times due to silicide structural relaxation. After annihilation of excess free volume a transition to parabolic growth takes place. Parameters of interdiffusion (activation energy $E \approx 1$ eV and pre-exponential factor $D_0 = 4\cdot10^{-12}$ m^{2}s^{-1}) were determined for parabolic growth stage.

Изучена кинетика фазообразования в многослойных периодических покрытиях Sc/Si и трехслойных структурах Si/Sc/Si методами электронной микроскопии поперечных срезов и малоугловой рентгеновской рефлектометрии. Рост силицида ScSi при всех температурах отжига лимитируется диффузией. Установлено две стадии роста силицида, подчиняющихся непараболическому и параболическому законам. Первоначальная непараболическая стадия роста характеризуется более высоким коэффициентом диффузии, который уменьшается в 20 раз в процессе изотермического отжига вследствие структурной релаксации структуры аморфного силицида. После аннигиляции свободного избыточного объема происходит переход к параболическому закону роста силицида. Стадия параболического роста характеризуется энергией активации $E = 1$ эВ и предэкспоненциальным множителем $D_0 = 4\cdot10^{-12}$ м^{2}с$^{-1}$.

Issues of diffusion and phase formation in transition metal/silicon layered systems are of great practical importance for up-to-date silicon technological applications in general and nanotechnologies in particular. Silicides of transition metals are nowadays widely used in semiconductor engineering, owing to their low specific electric resistances and elevated melting temperatures. These factors led researchers to undertake numerous experimental studies of diffusion processes and phase formation in metal-silicon systems [1, 2]. Furthermore, a number of multilayer periodic metal-silicon structures were found to be very perspective as reflecting components for X-ray and extreme ultraviolet (EUV) optics ($1 \le \lambda \le 50$ nm) due to their high experimentally reflection coefficients. As a good example, the Sc/Si periodic multilayers with periods of 20–35 nm have proven to be very perspective for application over the wavelength range of 35–50 nm [3]. Up to date, they are widely used to control the X-ray laser-radiation at $\lambda = 46.9$ nm [4], in X-ray microscopy [5] and in astrophysical research applications as well [6]. Optical properties of

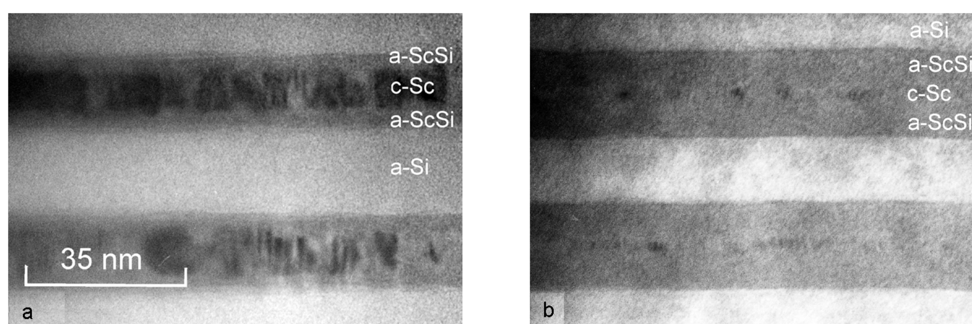

Fig. 1. Cross-sectional TEM images of the Sc/Si multilayers in as-deposited state (a) and after annealing at T_{ann} = 210°C for 24 hours (b). Si substrate (not shown) is below.

X-ray multilayers depend greatly on the presence of interlayers at interfaces between layers of basic materials. This fact became a next stimulus to study the processes of diffusion and phase formation in metal-silicon multilayer structures. In our previous work, we determined basic mechanisms of silicide formation in Sc/Si multilayers over the wide range of temperatures. Temperature interval of mirrors stability, type of chemical reactions and sequence of silicide formation were determined in papers [7–9], and the fast diffusion species was determined in [10]. The present paper refers to a detailed study of kinetics of silicide formation at initial stages which generally defines the thermal stability of X-ray Sc/Si multilayers.

Two types of samples were prepared: periodic Sc/Si multilayers consisting of thin layers and three-layer Si/Sc/Si coatings consisting of thicker layers. Period of multilayer (i.e., thickness of Sc/Si bilayers) was H = 35 nm with bilayer number amounted to N = 20. Thickness of every layer in the Si/Sc/Si layered composition was 100/277/100 nm, correspondingly. The multilayers were used for studying initial stage of silicide growth, while the later stage was studied using the three-layers (see below for details). All three-layers and multilayers were fabricated by method of dc magnetron sputtering in Ar environment under ~2.6 Pa working pressure. To study mechanisms and kinetics of silicide formation, the samples were subjected to isothermal annealing in a vacuum furnace at 10^{-2} Pa, over 130°C–400°C temperature range. After annealing for about 1 hour samples were cooled down and investigated by small-angle X-ray reflectivity (SAXR) techniques with primary focus to measure silicide thickness. The SAXR spectra were obtained in (θ–2θ) geometry at λ-$CuK_{\alpha1}$ = 0.154051 nm with DRON-3M diffractometer ("Burevestnik", Russia) equipped with a silicon monochromator. Experimental diffraction spectra were interpreted by comparison of them with simulated ones. The simulation of the diffraction spectra were performed using Fresnel equations [11–13]. In order to verify the results of simulation some test-samples were selectively studied by TEM techniques. Results of the thickness measurements obtained by both methods were in a good correlation. The cross-sectional TEM studies were performed in transmission electron microscope PEM-U ("SELMI", Ukraine) operated at 100 keV. The samples preparation procedure was described in paper [14].

The cross-sectional TEM image of the Sc/Si multilayer in as-deposited state is shown in Fig. 1a. There are amorphous interlayers of ~3 nm thick between the polycrystalline Sc and amorphous Si layers with thickness of 9.8 nm and 19.3 nm, correspondingly. The interlayers are some Sc_xSi_y silicide (alloy) formed during the deposition of the multilayer. During isothermal annealing at T_{ann} = 210°C, thickness of amorphous layers grew up at the expense of Sc and Si layers (Fig. 1b). Monitoring the change of Sc and Si thickness during annealing, the mean stoichiometric composition of a growing amorphous alloy can be estimated. For example: comparison of data in Fig. 1a and Fig. 1b shows that 6.2 nm Sc-layers and 4.9 nm Si-layers were consumed in relevant chemical reaction to form two silicide layers of near equal thickness of ≈4.5 nm at Sc-on-Si and Si-on-Sc interfaces. Thus, proportion of multilayer material consumption made up 1.27/1.0. The total thickness of each silicide layer, formed during deposition process and subsequent annealing amounted ≈7.5 nm. There are a few silicides in the binary Sc–Si phase diagram: Sc_5Si_3, Sc_5Si_4, ScSi, $ScSi_{1.22}$–$ScSi_{1.67}$ (i.e. Sc_4Si_5–Sc_3Si_5) [15]. The highest silicon content phase is widely non-stoichiometric.

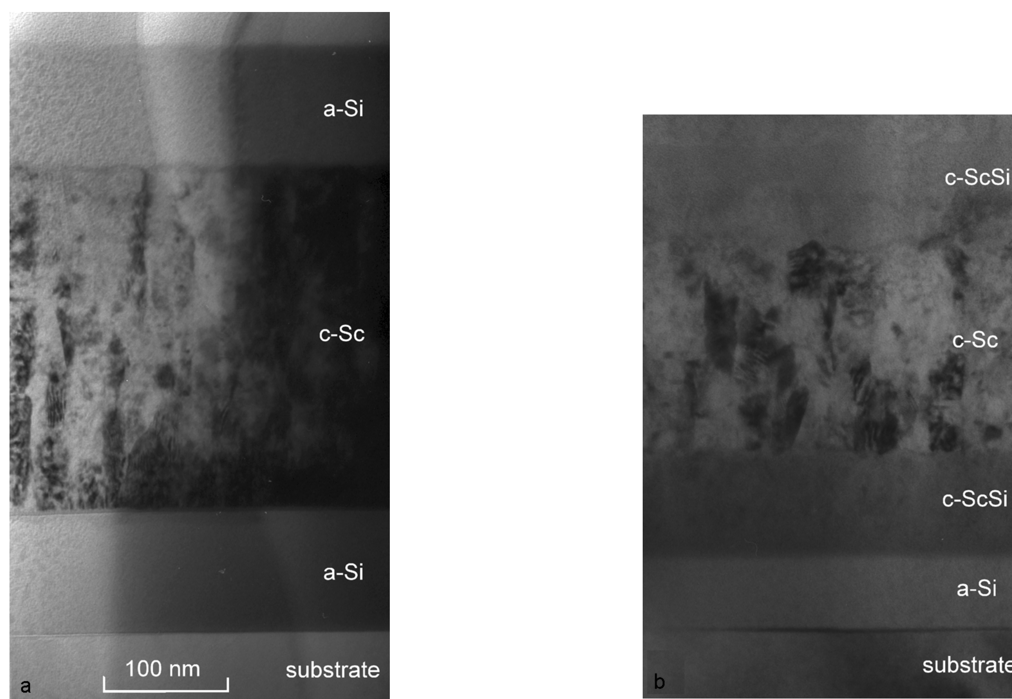

Fig. 2. Cross-sectional TEM images of the Si/Sc/Si three-layers in as-deposited state (a) and after annealing at T_{ann} = 400°C for 1 hour (b).

Known densities of the scandium silisides have close values ($\rho_{Sc_5Si_3}$= 3.299 g/cm3, ρ_{ScSi}= 3.363 g/cm^3, $\rho_{Sc_3Si_5}$ = 3.393 g/cm^3) [16].

We can calculate the thickness ratio of consumed scandium (Δt_{Sc}) and silicon (Δt_{Si}) at formation of some silicide Sc_mSi_n as:

$$\Delta t_{Sc}/\Delta t_{Si} = (m\rho_{Si}M_{Sc})/(n\rho_{Sc}M_{Si}), \quad (1)$$

where ρ_{Si} and ρ_{Sc} are densities (ρ_{Sc} = 3.02 g/cm^3 and ρ_{Si} = 2.33 g/cm^3) and M_{Si} and M_{Sc} are atomic masses of silicon and scandium, respectively, m and n are coefficients in chemical formula of a siliside.

Calculated ratios $\Delta t_{Sc}/\Delta t_{Si}$ made up 2.06, 1.54, 1.24 and 0.99–0.74 for the silicides Sc_5Si_3, Sc_5Si_4, ScSi, $ScSi_{1.22}$–$ScSi_{1.67}$ (Sc_4Si_5–Sc_3S_5), correspondingly. The experimental ratio of Sc and Si volume fractions (1.27/1.0) agrees well with theoretical proportion (1.24/1.0) in reaction of ScSi silicide formation. Thus, processes of silicide formation in Sc/Si multilayers, at temperatures up to 210°C, occurs under solid-state amorphization reaction, resulting in formation of amorphous ScSi: c-Sc + a-Si → a-ScSi. Further evolution of the structure and phase composition of Sc/Si layered systems (including transformations between amorphous silicides, crystallization and decomposition of crystalline silicides) was described in detail earlier [9].

The cross-section TEM images of Si/Sc/Si structures, in as-deposited state and after annealing at T_{ann} = 400°C for 1 hour, are shown in Fig. 2a and Fig. 2b. It is evident that ScSi formation in three-layer coatings runs in a manner similar to that in multilayers. Again, comparison of Fig. 2a and 2b shows that 54 nm of Sc and 43 nm of Si was consumed in this chemical reaction, resulting in formation of silicide layer 76 nm thick. It means that Sc and Si are consumed at the ratio of 1.26/1.0 and that composition of the silicide is the same for multilayers and three-layers and agrees well with the ScSi silicide stoichiometry. Thus, composition of the silicide at earlier and later stages is generally the same, and does not depend neither on the thickness of scandium and silicon layers nor the silicide layer thickness. Analysis of selected area diffraction pattern has shown that annealing at T_{ann} = 400°C for 1 hour results in formation of the crystalline ScSi silicide. The crystalline ScSi can also be revealed in the selected area diffraction pattern of the three-layer sample annealed at T_{ann} = 350°C for 11 hours. Note, that the thickness of the silicide, when crystalline phase was just detected, was ≈79 nm in both above cases. Thus, the amorphous silicide forms at in-

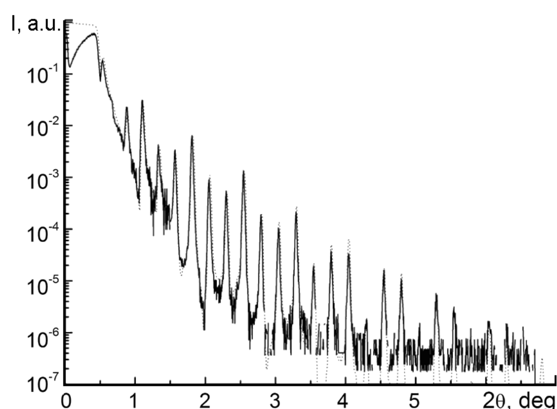

Fig. 3. Experimental (solid line) and calculated (dot line) small-angle X-ray (λ = 0.154 nm) spectra for the Sc/Si multilayers in as-deposited state.

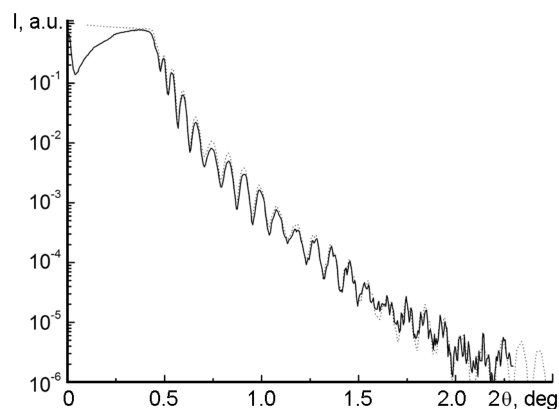

Fig. 4. Experimental (solid line) and calculated (drop line) small-angle X-ray (λ = 0.154 nm) spectra for the Si/Sc/Si three-layers after annealing at T_{ann} = 300°C for 1 hour.

itial stages of annealing and undergoes crystallization at further stages.

To plot a dependence of the silicide thickness on annealing time we used the SAXR data. Fig. 3 shows SAXR spectra of Sc/Si multilayers in as-deposited state. Up here, we can identify about 25 Bragg reflection orders from periodical structure of the multilayer. Angular position of peaks is defined by the Bragg's law, with account of X-rays refraction effect. Referring to the Bragg's law and plotting the $\sin^2\theta$ versus $n^2\lambda^2/4$ graph where n is order of Bragg peak, we can compute value of period in the multilayer with up to 0.05 nm accuracy allowance. The diffraction spectra contain, besides, the information on thickness of individual layers. This information can be extracted from the full set of Bragg peak intensities. In general case, the intensity of each Bragg peak is determined by interference of radiation beams reflected from all interfaces of the multilayer. As one can see in Fig. 1a there exist four types of interfaces in Sc/Si coatings, such as: Si-on-ScSi, ScSi-on-Sc, Sc-on-ScSi and ScSi-on-Si. At the same time, TEM imaging and SAXR spectra simulation techniques show that roughness of ScSi-on-Sc and Sc-on-ScSi interfaces is by far greater than that of Si-on-ScSi and ScSi-on-Si ones. Since rough boundaries of the Sc layer make a minor contribution to resulting diffraction, the intensity of Bragg peaks should mainly be defined by position of ScSi-on-Si and Si-on-ScSi interfaces, i.e. by Si layer thickness or, more exactly, by ratio of Si layer thickness to the period.

Knowing composition of the growing silicide and Si layer thickness evolution, we can predict the change of silicide thickness during annealing, because both these values are correlated together by ratio:

$$\Delta h_{Si}/(2 \cdot \Delta h_{ScSi}) = 0.554. \quad (2)$$

This correlation is determined by the ratio of molar volumes of both components in Sc + Si→ScSi chemical reaction, where Δh_{Si} denotes reduction of Si thickness, Δh_{ScSi} defines increment of silicide interlayer thickness; and coefficient 2 accounts for existence of two silicide interlayers in each period. The silicide formation reaction is accompanied by change of specific volume as:

$$\Delta H = \Delta h_{Sc} + \Delta h_{Si} - 2 \cdot \Delta h_{ScSi}, \quad (3)$$

where ΔH is the change of the multilayer period and Δh_{Sc} denotes the reduction of Sc thickness. Hence, evolution of silicide thickness can also be evaluated by change of the multilayer period (ΔH) as:

$$\Delta h_{ScSi} = 2.09 \cdot \Delta H. \quad (5)$$

The SAXR diffraction curves for Si/Sc/Si three-layers in as-deposited state and after annealing at 300°C for 1 hour are shown in Fig. 4. Character of intensity oscillations for diffraction angles $2\theta<2°$ is determined mainly by thickness of the top silicon layer (defined with accuracy of ~0.5 nm). Value of silicide thickness can be calculated from the Si consumption per ratio:

$$\Delta h_{Si}/\Delta h_{ScSi} = 0.554. \quad (5)$$

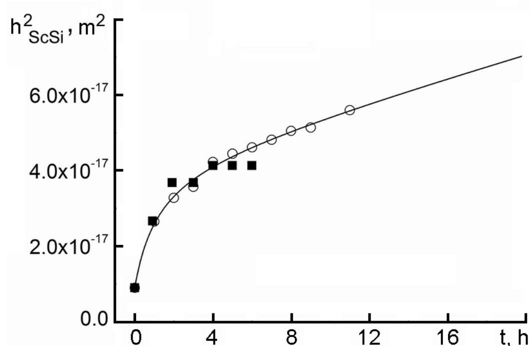

Fig. 5. Time dependences of squared thickness of the silicide ScSi (h^2 $_{ScSi}$) in the multilayers on duration of isothermal annealing process at T_{ann} = 210°C. The fitting curves are shown as solid lines. The dark squares (■) in the Figure correspond to the thee-layer annealed at the same temperature.

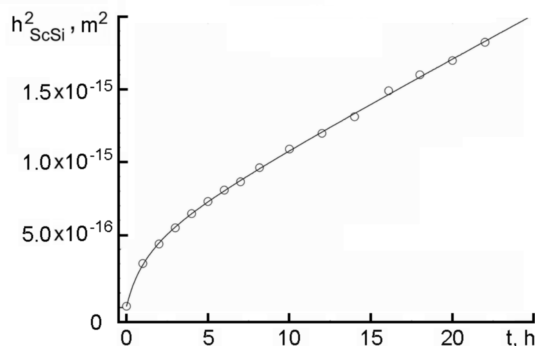

Fig. 6. Time dependences of squared thickness of the silicide ScSi (h^2 $_{ScSi}$) in the Si/Sc/Si three-layers on duration of isothermal annealing process at T_{ann} = 300°C. The fitting curves are shown as solid lines.

Fig. 5 represent dependences of amorphous silicide thickness on annealing time $h^2(t)$ for multilayers treated at temperatures of 210°C. It is obvious that character of the silicide growth is rather complicated. On one hand, dependence of the thickness on time does not manifest its linear character ($h \sim t$), that means this process does not obey the boundary kinetics. In other words, process of silicide growth is not governed by a chemical reaction of scandium and silicon at the interfaces but is limited by rate of diffusion of Si or/and Sc atoms through the silicide layer. On the other hand, silicide growth does not obey the parabolic law ($h^2 \sim t$) at least at initial stage as it should be in the case of diffusion kinetics. The dependence $h^2 \sim t$ becomes linear only after long annealing at moderate temperatures (Fig. 5). In order to provide a correct comparison of results collected from multilayers and three-layers, it is necessary to check out whether silicide growth rates are equal in both cases and whether any size-effects appear. Time-dependence of silicide layer thickness in three-layers at 210°C is shown in Fig. 5 with dark squares. The scatter in the three-layer data is much broader than that in multilayer data (open circles) because less precision of layer thickness measurement for three-layers (Fig. 5). Nevertheless, both dark squares and open circles display a fairly good mutual agreement along the joint characteristic curve for both three-layers and multilayers. Therefore, silicide growth rate does not depend on thickness of Sc layers. Hence, comparison of results collected on three-layers and multilayers can be regarded to as a quite correct procedure. Dependences of silicide growth on annealing time for three-layers treated isothermally at 300°C are presented in Fig. 6. The plots show quite clearly that after initial stage of non-linear growth, a transition to linear growth stage occurs.

According to classic theory of reactive diffusion a growth of an intermediate compound occurs as a result of two successive processes [17]. The first one is a diffusion of reaction-involved components through the layer of intermediate compound toward the reaction zone. Kinetics of this process is described by parabolic law ($x^2 \sim t$), where x is diffusion length (i.e. thickness of the intermediate compound) and t is annealing time. The second process is a chemical reaction that takes place at boundaries of the intermediate compound layer at a constant time-independent rate. At initial stage of growth, when thickness of the intermediate compound is small (smaller than some transient thickness x^*), the diffusion runs very fast, because its rate is inversely proportional to thickness as $dx/dt \sim 1/x$. At this stage, chemical reaction is the slowest process that specifies the constant rate ($x \sim t$) of thickness growth. However, as long as the silicide thickness is growing on, diffusion rate is decreasing, and the diffusion becomes the slowest process when thickness exceeds the value x^*. Under these conditions, silicide growth kinetics changes towards a diffusion regime ($x^2 \sim t$). Results of this research show, however, that character of amorphous silicide growth differs substantially from the classic behavior. The stage of boundary growth kinetics of ($h \sim t$) is not observed. The section of the curve

corresponding to the boundary kinetics is absent in the Fig. 5 and 6. In terms of $h^2(t)$ coordinates this imaginary section should look like an uprising curve. Thus, absence of such an uprising curve-section at the very beginning (or, at least, when silicide layer thickness exceeds 3 nm) indicates that the silicide growth process is limited by the diffusion only and not by the chemical reaction. In other words, value of transient thickness x^* for the Sc/Si system is likely to be substantially below 3 nm.

Secondly, on initial stage of growth, the parabolic law ($h^2 \sim t$) is disobeyed, because of dependence of squared thickness of the silicide layer on time is not linear. In other words, the effective diffusion coefficient is not constant, but decreases during the annealing as the angle of slope for curves $h^2(t)$. For example, the diffusion coefficient was $\sim 2 \cdot 10^{-21}$ m^2s^{-1} for the first hour, $\sim 0.8 \cdot 10^{-21}$ m^2s^{-1} for the second hour and $\sim 0.3 \cdot 10^{-21}$ m^2s^{-1} for the eleventh hour of annealing at 210°C. Below in this paper, there is a discussion on possible reasons that bring to slowdown of the silicide growth, meaning such aspects as non-linear diffusivity and structural relaxation of amorphous layers during annealing. At later stages, there occurs transition to regime of classic reactive diffusion, whereby dependence of squared thickness (h^2) on time (t) becomes linear. The similar slowdown of an amorphous silicide growth was observed in due time in Mo/Si multilayers [18, 19].

Keeping in mind that diffusion is going on through the ultra-thin layers, size-effects have to be taken into consideration. Having analyzed a case of reactive diffusion through a very thin intermediate compound layer, author of the paper [21] concluded that the law of diffusion-flow proportionality to concentration-gradient ($J \sim dC/dx$), known as the first Fick's law, would no longer be obeyed if values of thickness fall below ~10 nm. In regard to low thickness values, the diffusion driving force (i.e. concentration gradient) becomes dramatically large. In that case the expression for a diffusion flow along with linear term (dC/dx) contains also terms of higher order like $(dC/dx)^3$ and more, which contribution is inversely proportional to the thickness x. In other words, with low values of thickness, dependence of diffusion flow on the concentration gradient is very strong and non-linear, whereby mass-transfer processes becomes highly accelerated. In a non-linear diffusion model, there must theoretically exist a certain critical thickness of transition toward the linear diffusion. It should be noted here that this model does not suggest any dependence of the critical thickness on temperature of annealing. However, judging by the situation in Fig. 5 and 6, thickness of transition to the linear stage of diffusion is different for the different temperatures. One has to take into account that the coatings were synthesized under far non-equilibrium conditions (such as quenching of vapor phase on a cold substrate). As a result the coatings contain lots of non-equilibrium vacation-type defects, excessive free volume and so on [21]. Such kind of defects facilitates the diffusion of silicon and scandium atoms through the silicide layers. However the defect concentration decreases during annealing due to annihilation of excessive free volume in layers of amorphous ScSi, and the diffusion slows down. This approach was applied particularly in paper [22] to analyze the kinetics of silicide growth in Mo/Si multilayer system. According to the approach there is an additional term $D_{nonequilib}$. Of diffusion coefficient describing the diffusion through non-equilibrium defects:

$$D = D_{equilib} + D_{nonequilib} \tag{6}$$

$$D_{nonequilib} = \frac{1}{(1/D_{initial} + d(1/D_{nonequilib})/dt * t)}. \tag{7}$$

The longer the annealing time, the smaller value of non-equilibrium term ($D_{nonequilib.}$) is; eventually it becomes negligible.

The presence of the non-equilibrium term of the diffusivity transforms the parabolic law of growth ($h^2 = 4Dt$) into:

$$h^2 = 4\left(\frac{1}{\frac{1}{D_{initial}} + d(\frac{1}{D_{nonequilib}})/dt \cdot t} + D_{equilib.}\right) \cdot t. \tag{8}$$

Taking into account the thickness of silicide layer before annealing, h_0 is not zero, we finally obtain the relationship between h and t for silicide growth:

$$h^2 - h_0^2 = = 4 \cdot \left(\frac{1}{\frac{1}{D_{initial}} + d(\frac{1}{D_{nonequilib}})/dt \cdot t} + D_{equilib}\right) \cdot t \tag{9}$$

This equation (9) was used to approximate experimental data on the silicide growth. The fitting curves are shown in Fig. 5 and 6 as thin continuous lines. This

procedure allows one to determine correctly the equilibrium diffusivity (i.e. the diffusion coefficient at parabolic stage of silicide growth at $D_{equilib.} = D_{parabolic}$) and to estimate the diffusivity at initial moment of annealing ($D_{initial}$) as well. Resultant approximation curves are plotted in Fig. 5 and 6 with thin continuous lines. As fitting parameters, there were used coefficients of diffusion at parabolic stage of growth ($D_{parabolic}$) and those at initial moment of annealing ($D_{initial}$) at the annealing time $t_{ann.} = 0$. Plots of Arrhenius dependencies for $D_{parabolic}$ and $D_{initial}$ are shown in Fig. 7. The activation energy E of diffusion on parabolic section of silicide growth curve and the pre-exponential factor makes up 1.0 eV and $D_0 = 4\cdot10^{-12}$ m^2s^{-1} correspondingly.

Earlier using diffusion marks method, we have found that the silicon is the fast diffusion species in Sc–Si system [9]. The intrinsic diffusivity of silicon in ScSi was found to exceed one of scandium by factor of 20. Therefore, the diffusion parameters involved (i.e. activation energy and pre-exponential factor) are relevant to diffusion of Si atoms through the amorphous ScSi silicide. Arrhenius plots yield low values of activation energy (E = 1.0 eV) and pre-exponential factor ($D_0 = 4\cdot10^{-12}$ m^2s^{-1}) for the diffusivity. Such the low values of diffusion parameters are typical for many amorphous alloys, thus confirming the known models on collective mechanisms of diffusion in amorphous materials [23]. In our research the scandium silicide was amorphous at all stages of annealing of multilayers and at initial stage of annealing of three-layers. The crystallization of amorphous silicide was observed at the later stages of annealing of tree-layers (T_{ann} = 400°C, t_{ann}≤1 hour and T_{ann} = 300°C, t_{ann}≤11 hours). We did not observe any noticeable changes of the activation energy during amorphous-to-crystalline transition. The most probable explanation of this fact may be related to a poor perfection of the silicide structure being formed under moderate temperatures of annealing. In the case the diffusion along the numerous defects of nanocrystalline silicide, such as dislocations and grain boundaries, plays the basic role. As the activation energy of diffusion along the crystalline structure defects is substantially lower as compared to that in volume of a perfect crystal, the activation energy of diffusion in the nanocrystalline silicide remains rather small.

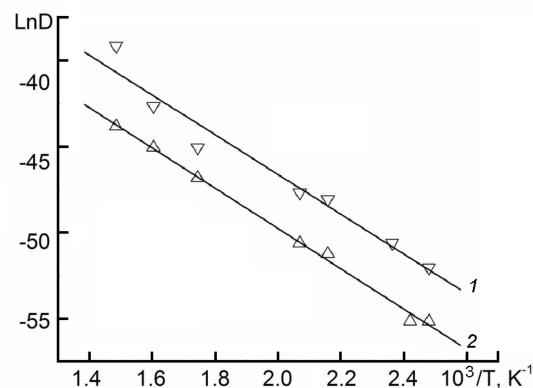

Fig. 7. Arrhenius plots for diffusion of Si atoms through the silicide ScSi at the initial stage *1* — (non-parabolic law) and at late stage *2* — (parabolic law) of silicide growth: E = 1eV, $D_0 = 3.75\cdot10^{-12}$ m^2s^{-1}.

ScSi silicide is determined to form in Sc/Si layered systems annealed over the 130–400°C temperature range, as a result of the chemical reaction: c-Sc + a-Si → a(c)-ScSi. At initial stage of growth, the scandium silicide is amorphous. As soon as the silicide layer is thick enough, the crystallization of the amorphous ScSi occurs (T_{ann} = 400°C, t_{ann}≤1 hour and T_{ann} = 300°C, t_{ann}≤11 hours). The silicide composition and its growth rate do not depend on thickness of Sc and Si layers. Growth of the silicide from the very beginning (x≥3 nm) obeys to diffusion-controlled kinetics. The stage of reaction-governed (h~t) kinetics was not revealed. Diffusion of Si atoms through the silicide layer is characterized by low values of activation energy (1.0 eV) and pre-exponential factor ($D_0 = 4\cdot10^{-12}$ m^2s^{-1}). The initial stage growth of amorphous silicide has an explicit non-linear behavior and the parabolic law h^2~t is disobeyed. The stage is characterized by decrease of diffusion coefficient by factor of 15–20 due to structural relaxation of amorphous silicide. At the isothermal annealing the transition to the classic parabolic law of the silicide growth occurs as annealing time increases.

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Початкові стадії дифузії і фазоутворення в шаруватих системах Sc/Si

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Вивчено кінетику фазоутворення у багатошарових покриттях Sc/Si і тришарових структурах Si/Sc/Si методами електронної мікроскопії поперечних зрізів і рентгенівської малокутової рефлектометрії. Зростання товщини силіциду ScSi при всіх температурах відпалу лімітується дифузією. Встановлено дві стадії росту силіциду, які відповідають непараболічному і параболічному законам. Первинна непараболічна стадія росту характеризується більшим коефіцієнтом дифузії, який зменшується у 20 разів у процесі ізотермічного відпалу як наслідок структурної релаксації структури аморфного силіциду. Стадія параболічного росту характеризується енергією активації $E = 1$ еВ і передекспоненціальним множником $D_0 = 4{\cdot}10^{-12}$ м^{2}с$^{-1}$.